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Title: Nuclide Identification, Quantification, and Uncertainty

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Nuclear Forensics Technical Measurement Training

- Gamma Spectrometry -

Nuclide Identification, Quantification, and Uncertainty

Magurele, Romania

Date: after Corona

Objective

The objective of this presentation is to provide an overview of nuclide identification, quantification, and uncertainty using gamma-ray spectrometry.

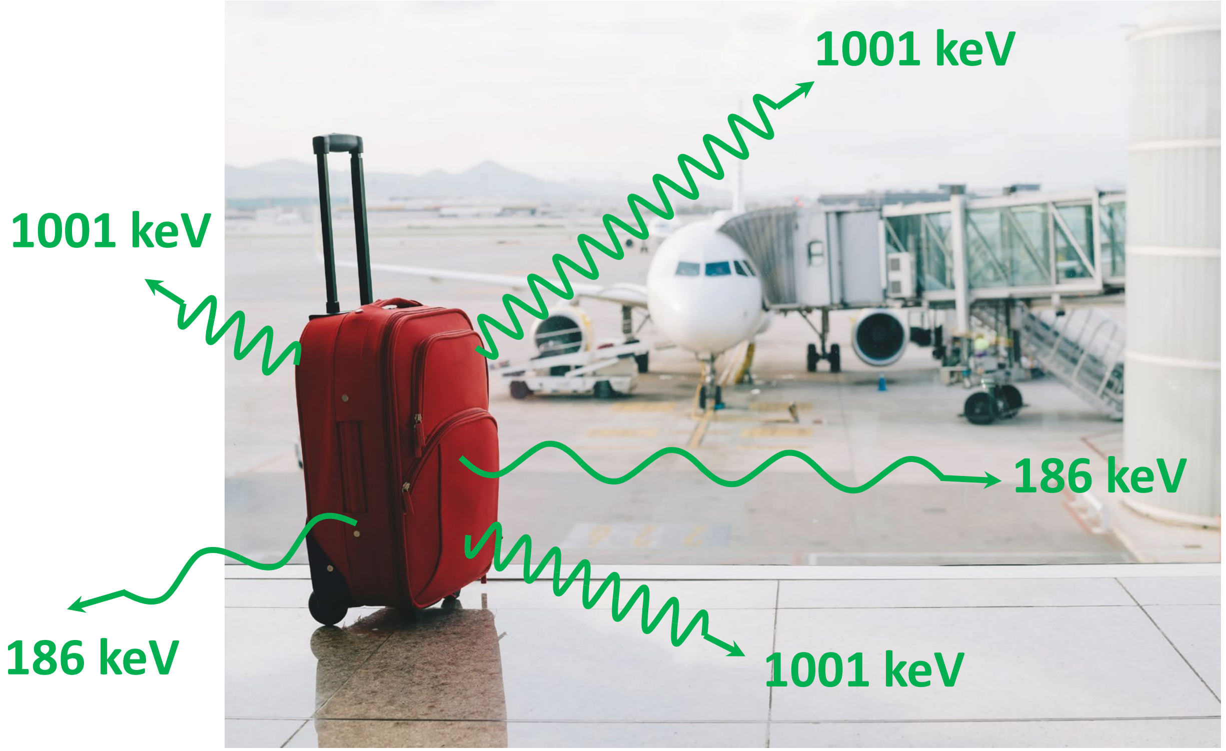
Part 1: Nuclide Identification

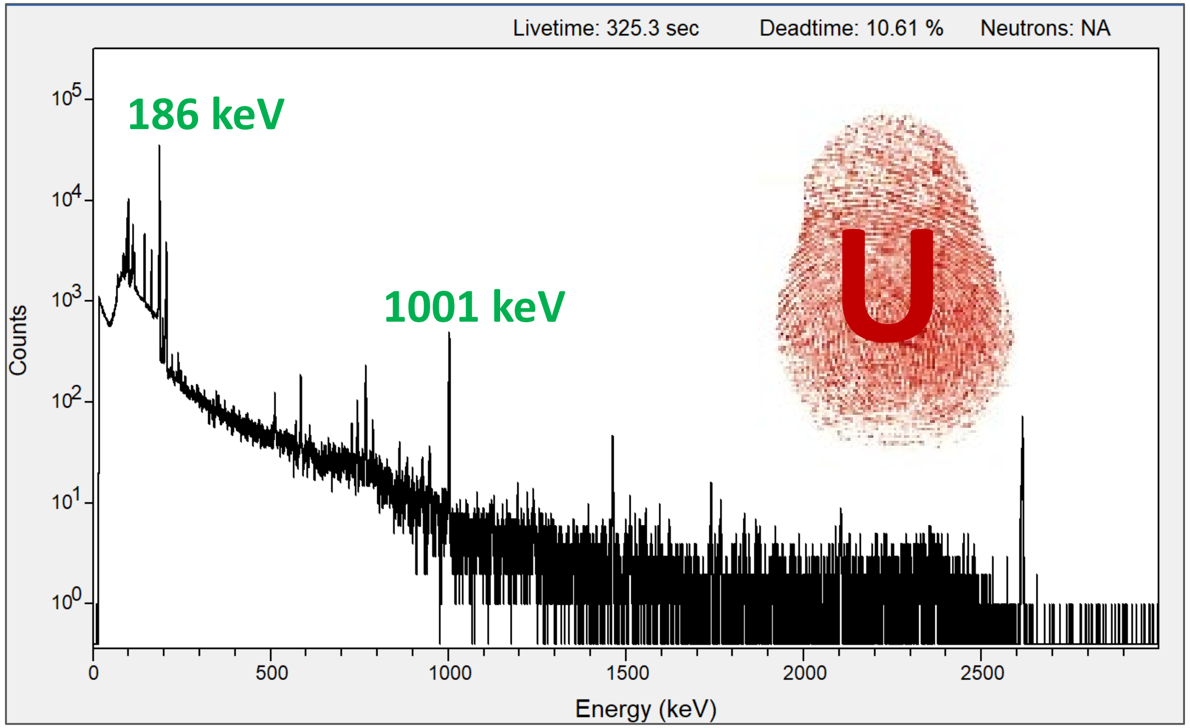
The first step in a gamma-ray analysis of an item is to determine what gamma-emitting nuclides are present in the item.

Gamma-ray spectrometry can be used for relatively rapid, initial, non-destructive identification of radionuclides in an item of interest before destructive analysis methods are employed.

Gamma Rays as "Fingerprints for Radionuclides"

- Gamma rays are specific to a radionuclide

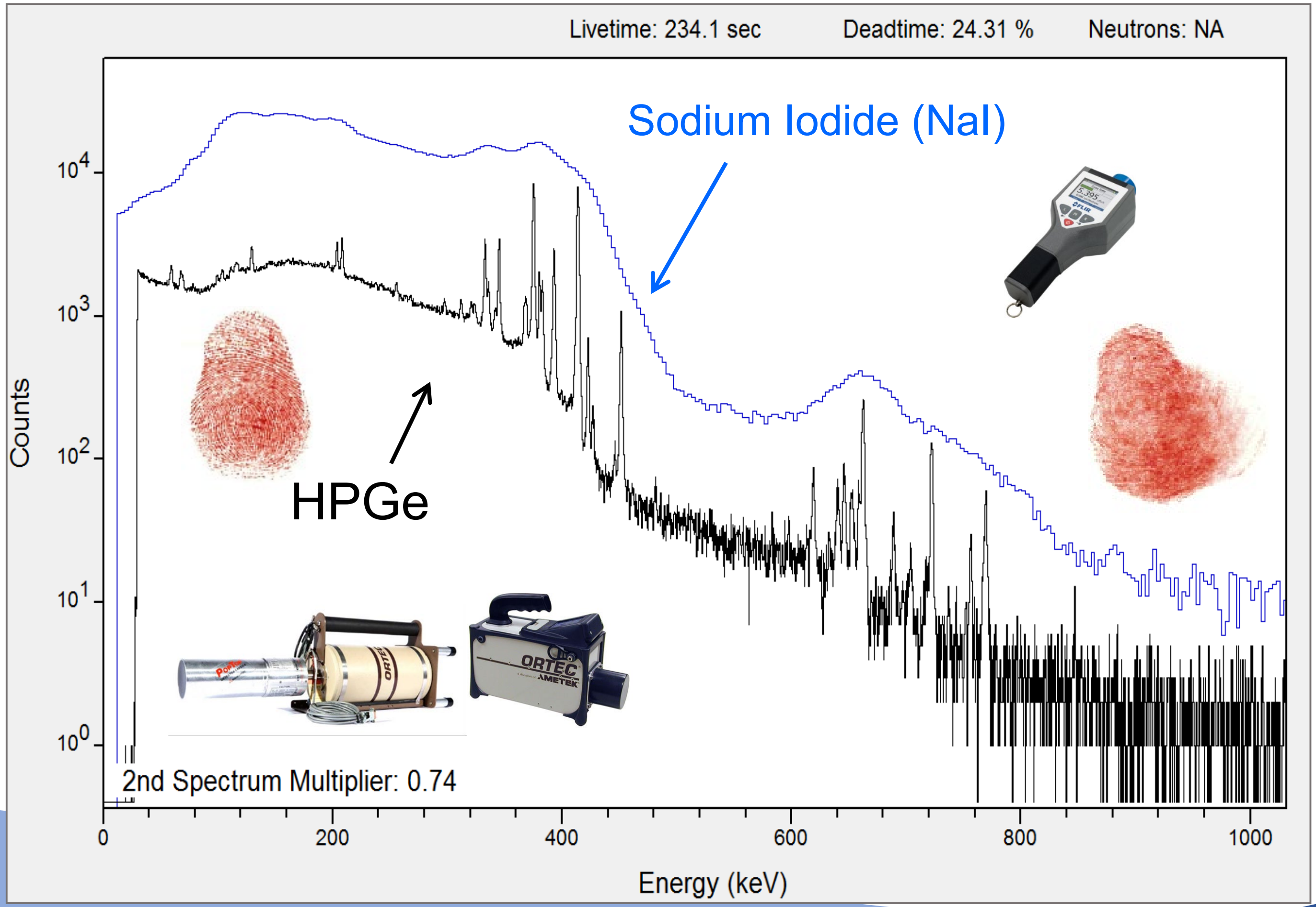




<u>Gamma-Ray Spectrum</u>: Histogram of energies deposited in the detector.

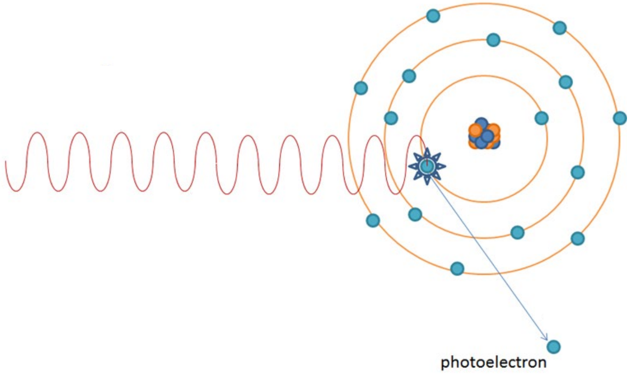
Gamma Rays as “Fingerprints for Radionuclides”

- The observed gamma-ray signature can change with measurement conditions such as shielding or detector type.



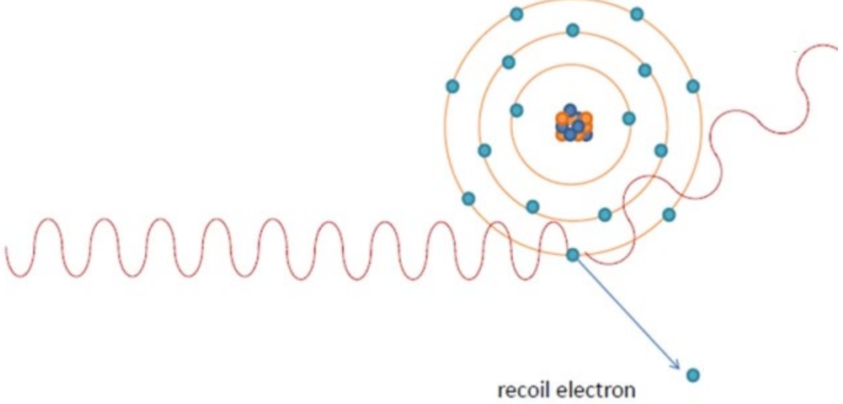
Photon Interactions with Matter

Photoelectric Effect



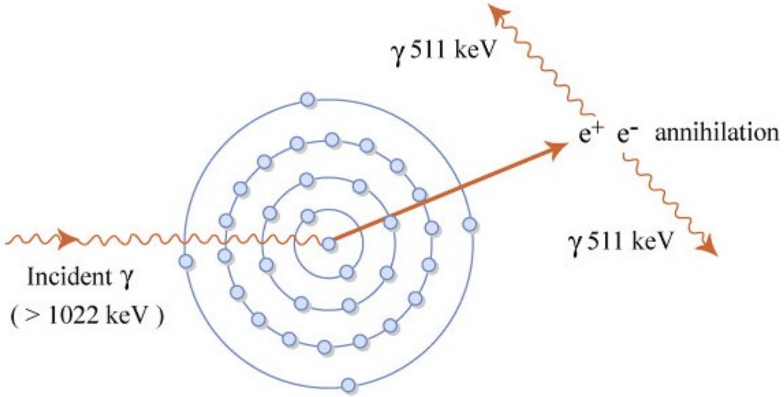
- Required to measure full-energy peaks
- Also contributes to continuum

Compton Scattering



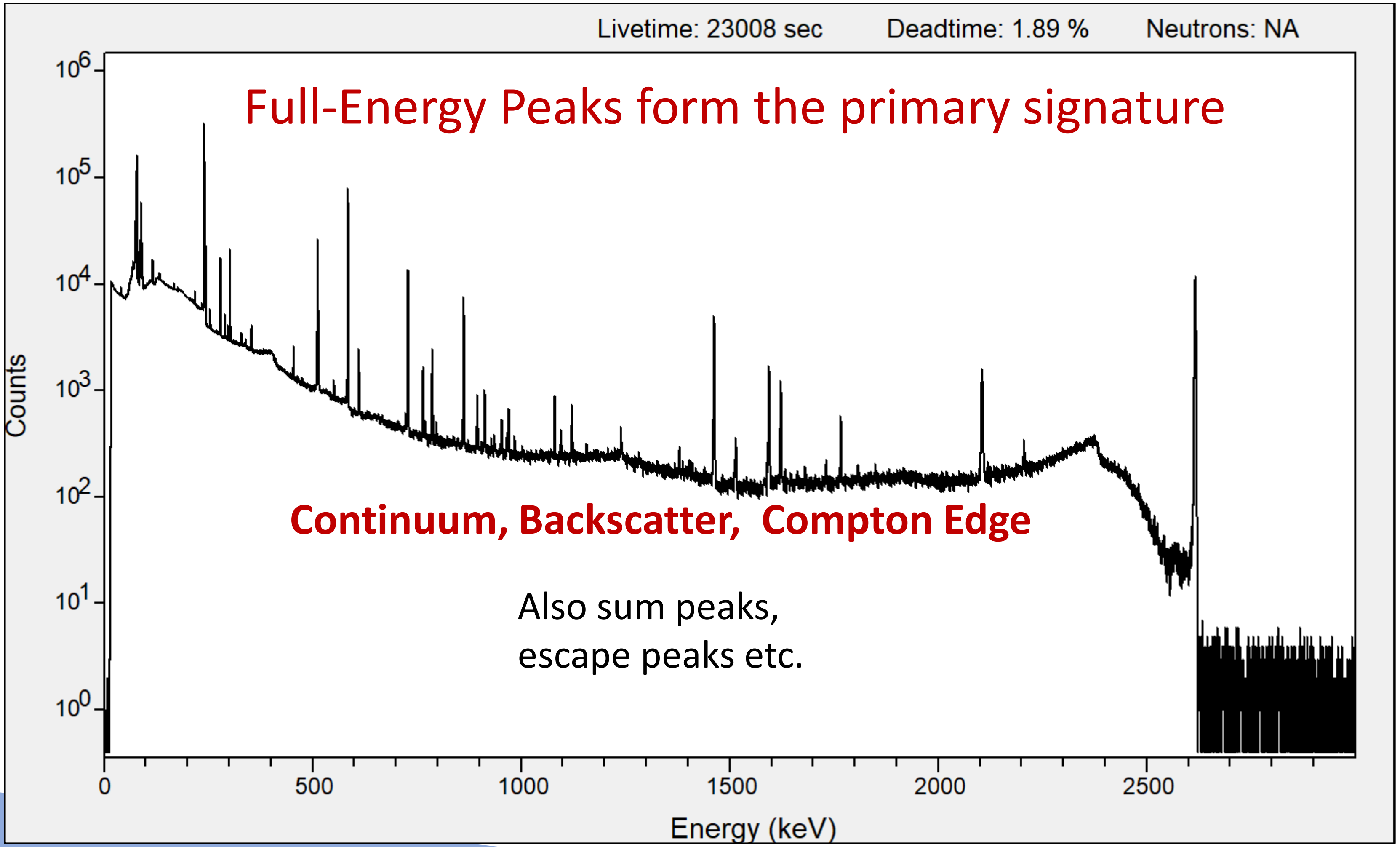
- Origin of the continuum
- Also contributes to full-energy peaks

Pair Production and Annihilation



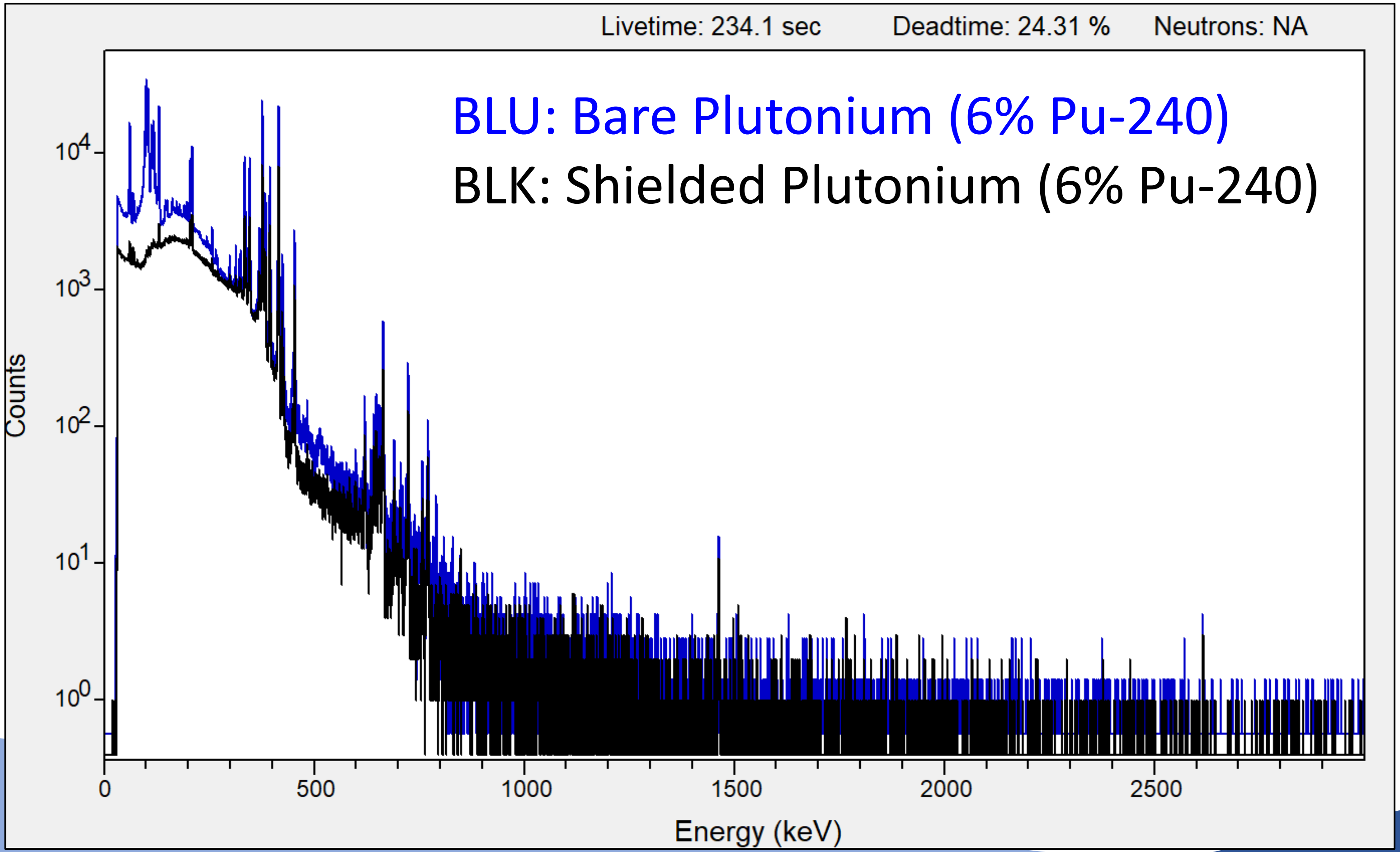
- Single and double escape peaks
- Annihilation radiation
- Also contributes to full-energy peaks

How do Photon Interactions Affect the Data?



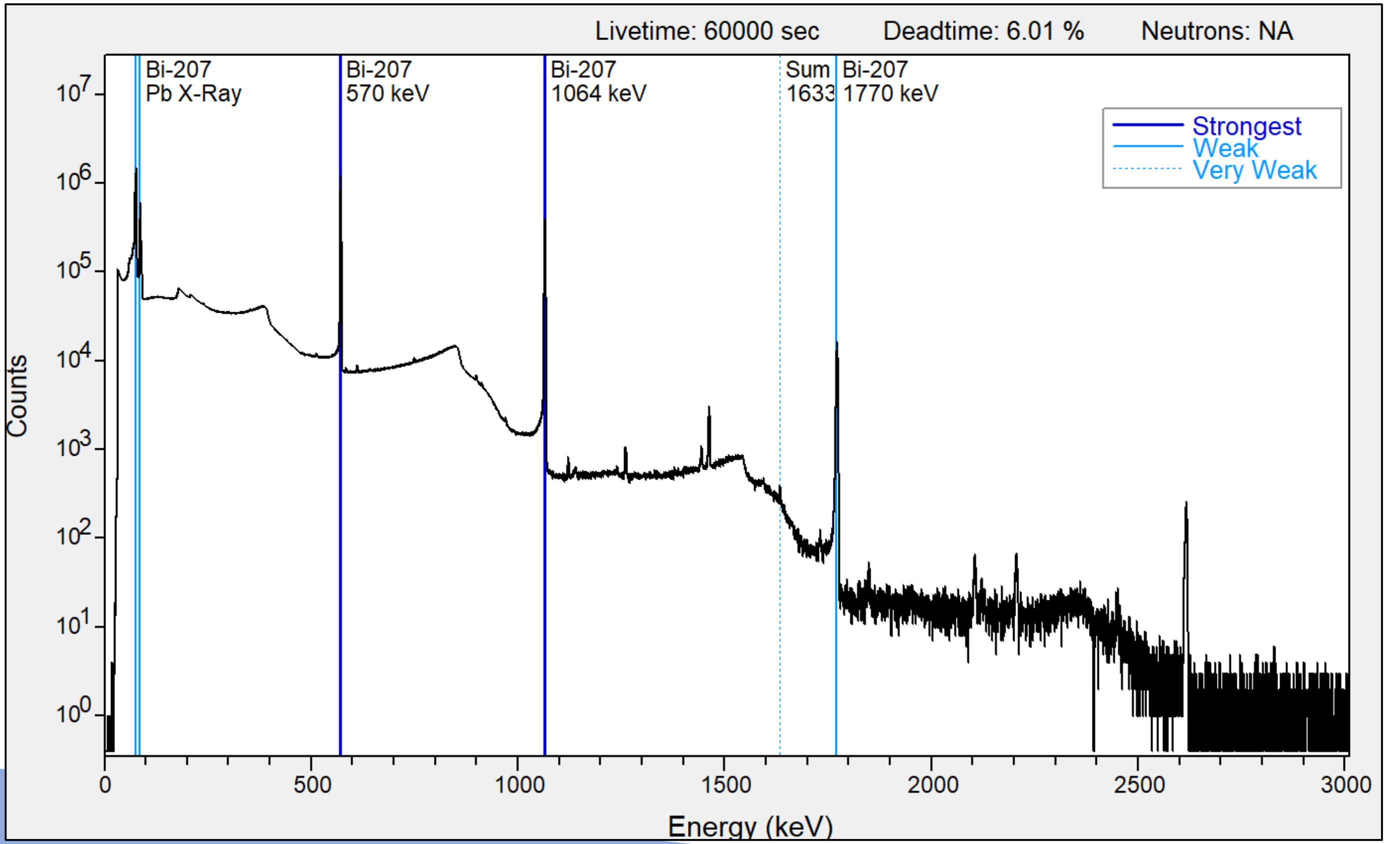
Using Pattern Recognition

- Pattern recognition can help expedite analysis
- Different measurement conditions can change the observed pattern

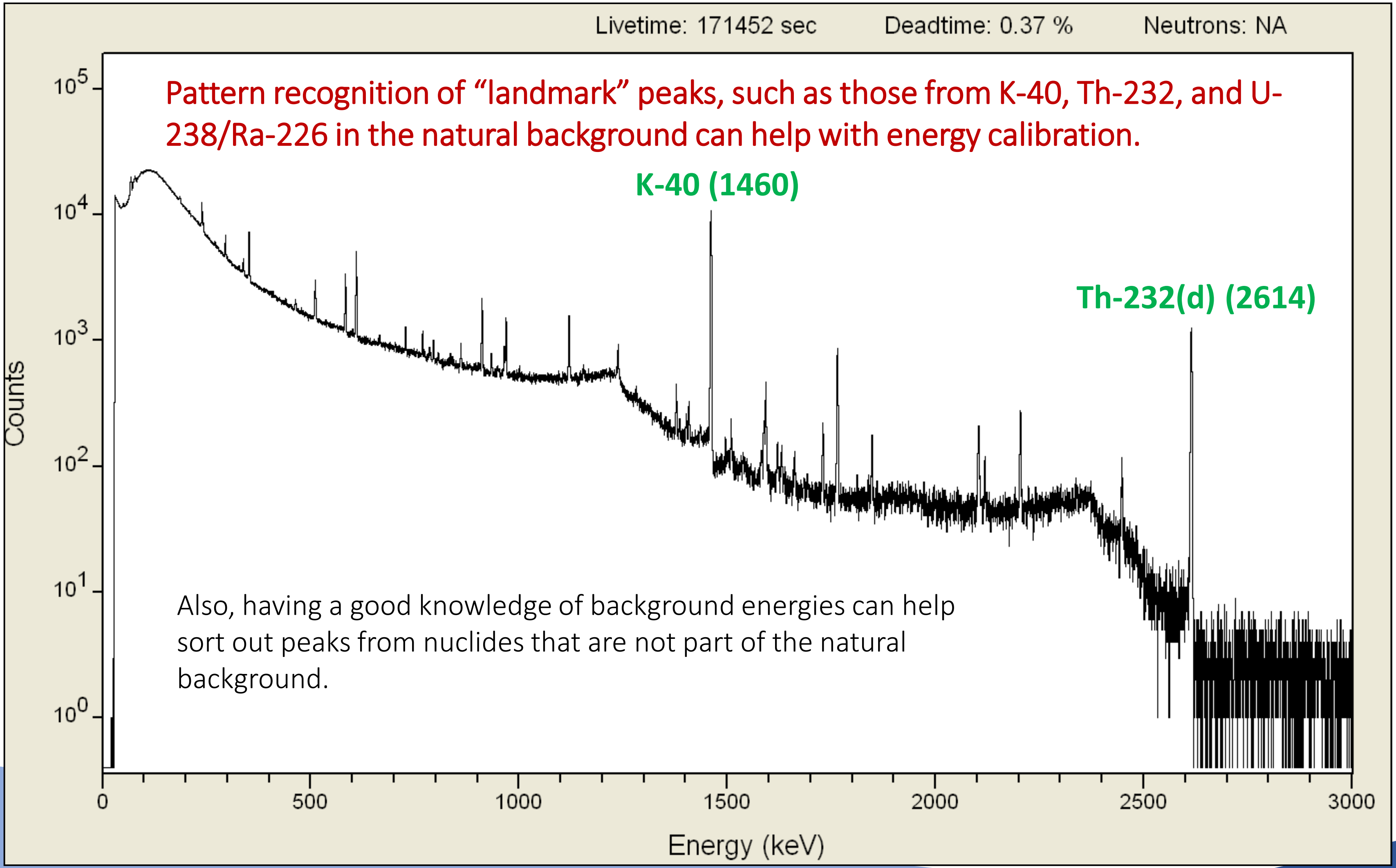


Using an Energy-Based Search

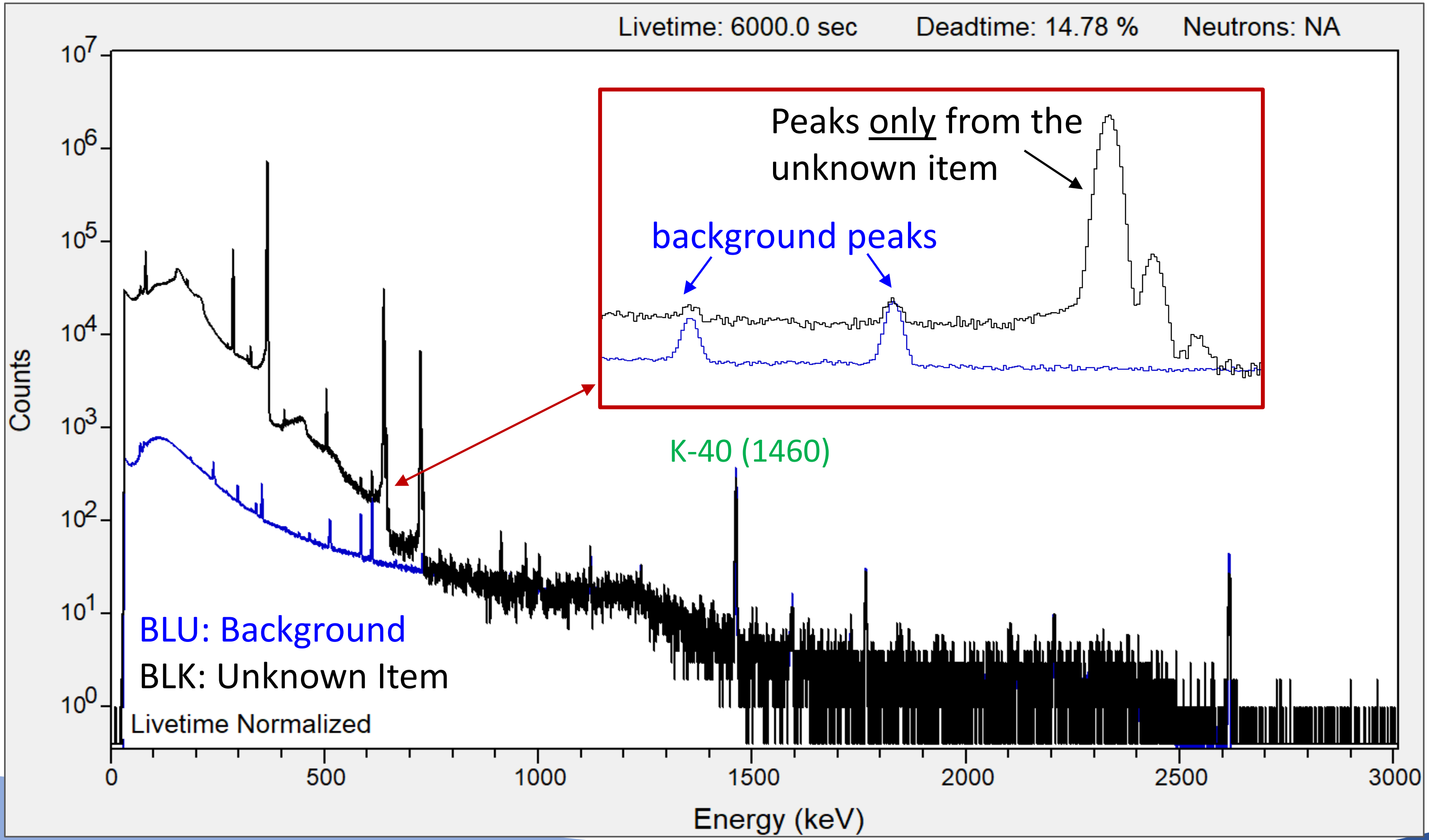
- An energy-based confirmation of nuclide identification should always be used
- You must have a good energy calibration to do this.



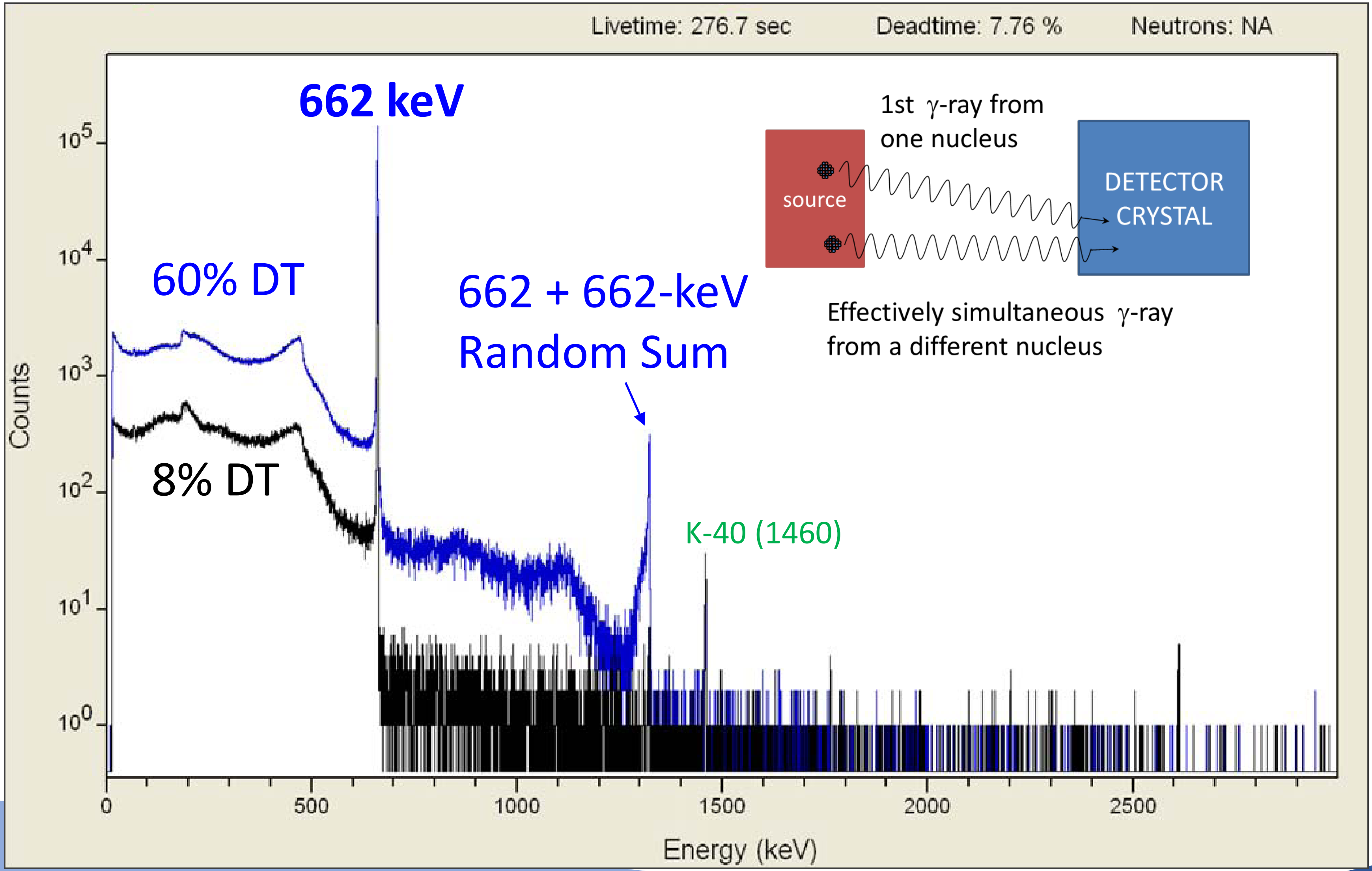
Understand the Natural Background



Comparing your Unknown Item to Background

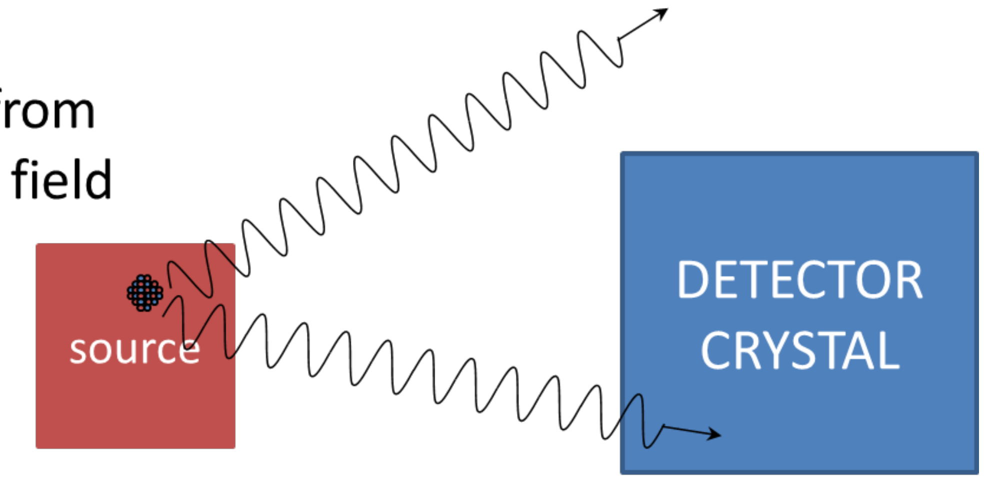


Other Spectral Effects: Random Coincidence

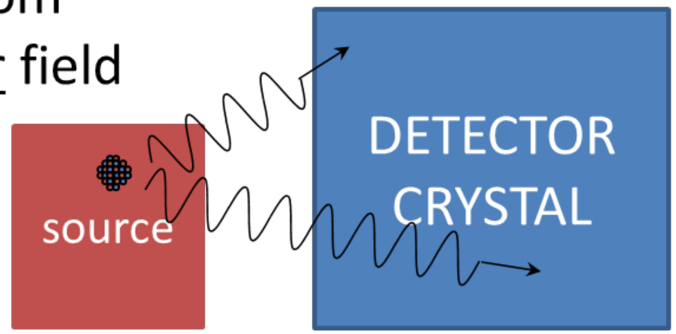


Other Spectral Effects: True Coincidence

2 simultaneous γ-rays from
<u>same</u> nucleus in the <u>far</u> field

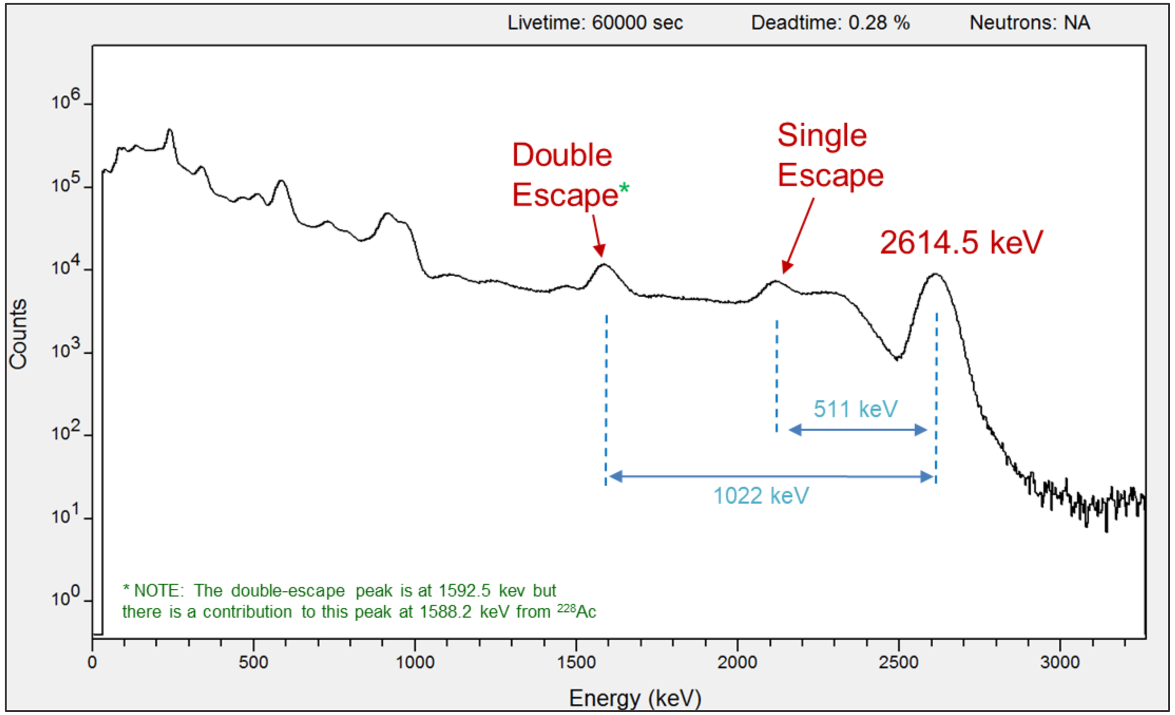


2 simultaneous γ-rays from
<u>same</u> nucleus in the <u>near</u> field

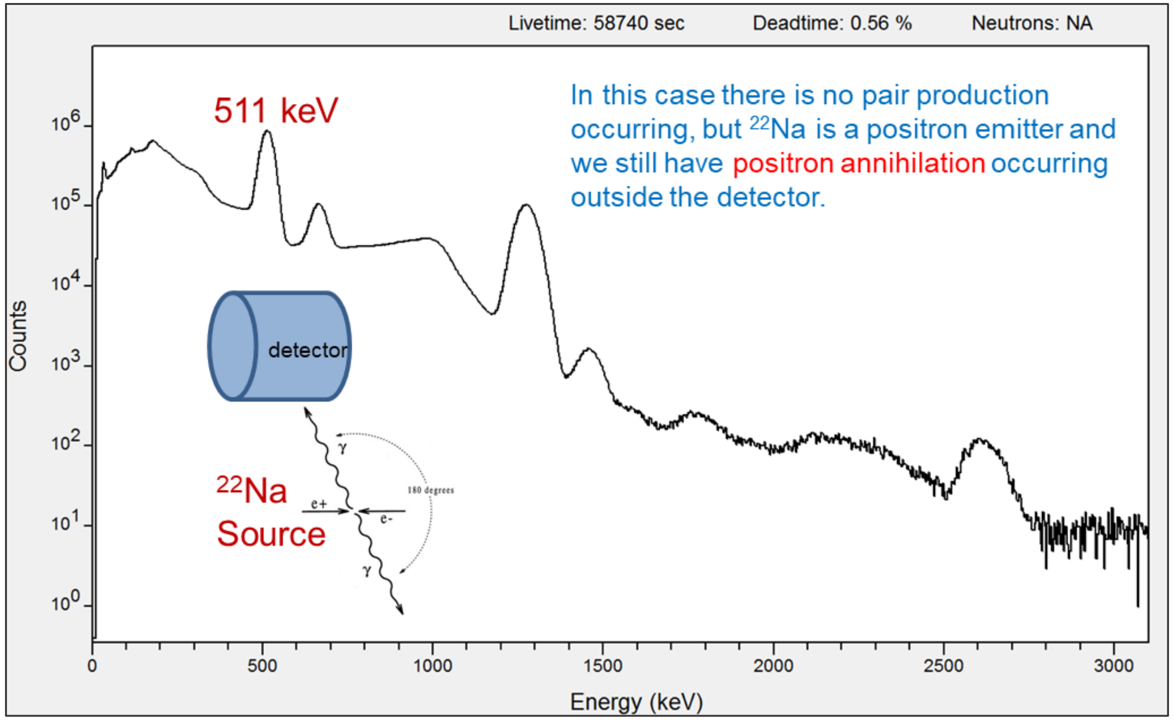


Escape Peaks & Annihilation Radiation

Escape Peaks

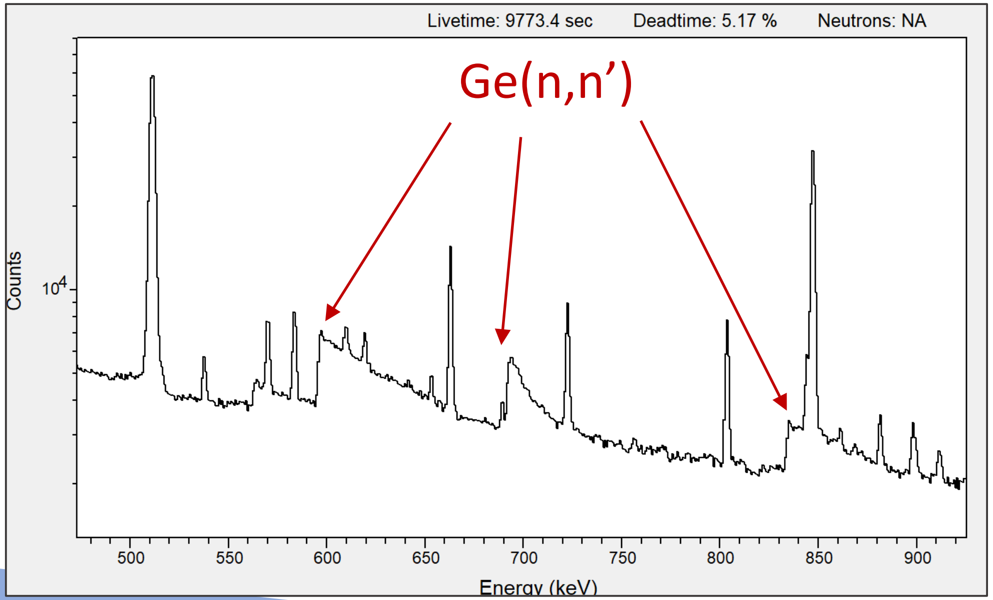


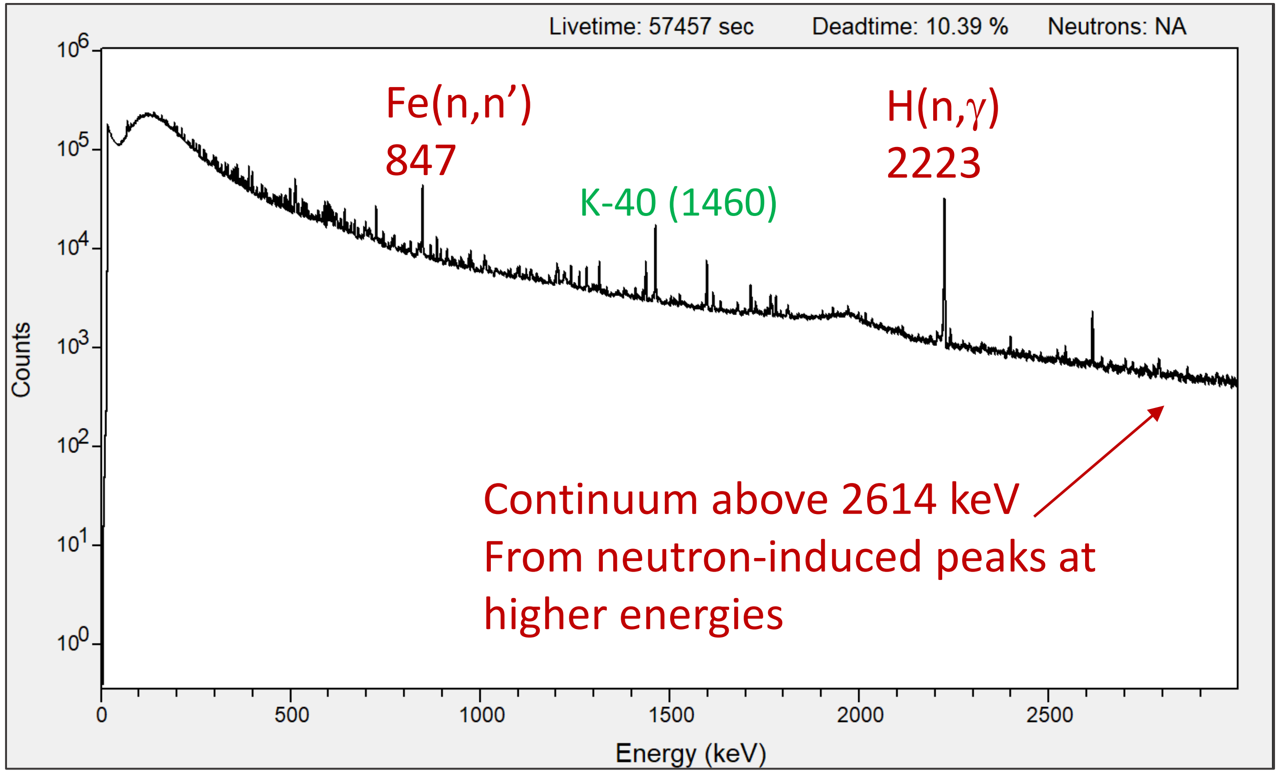
Annihilation Radiation



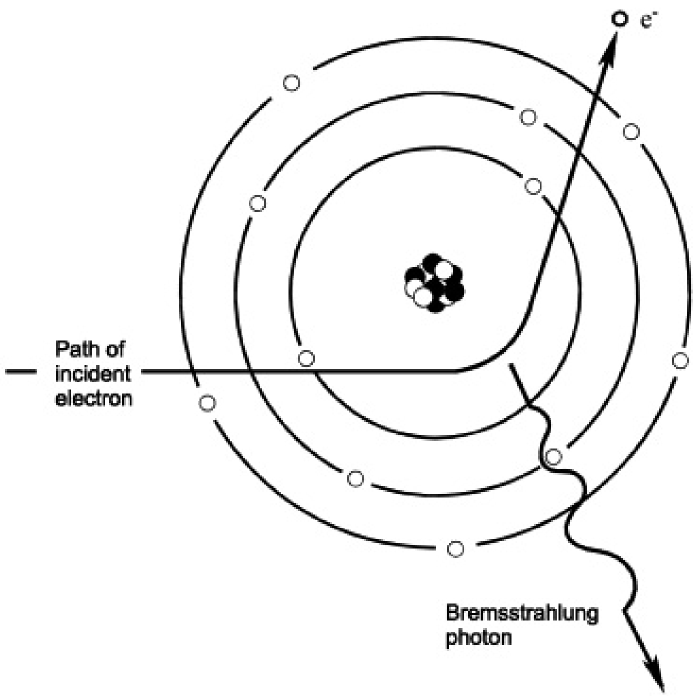
Neutron-Induced Gammas

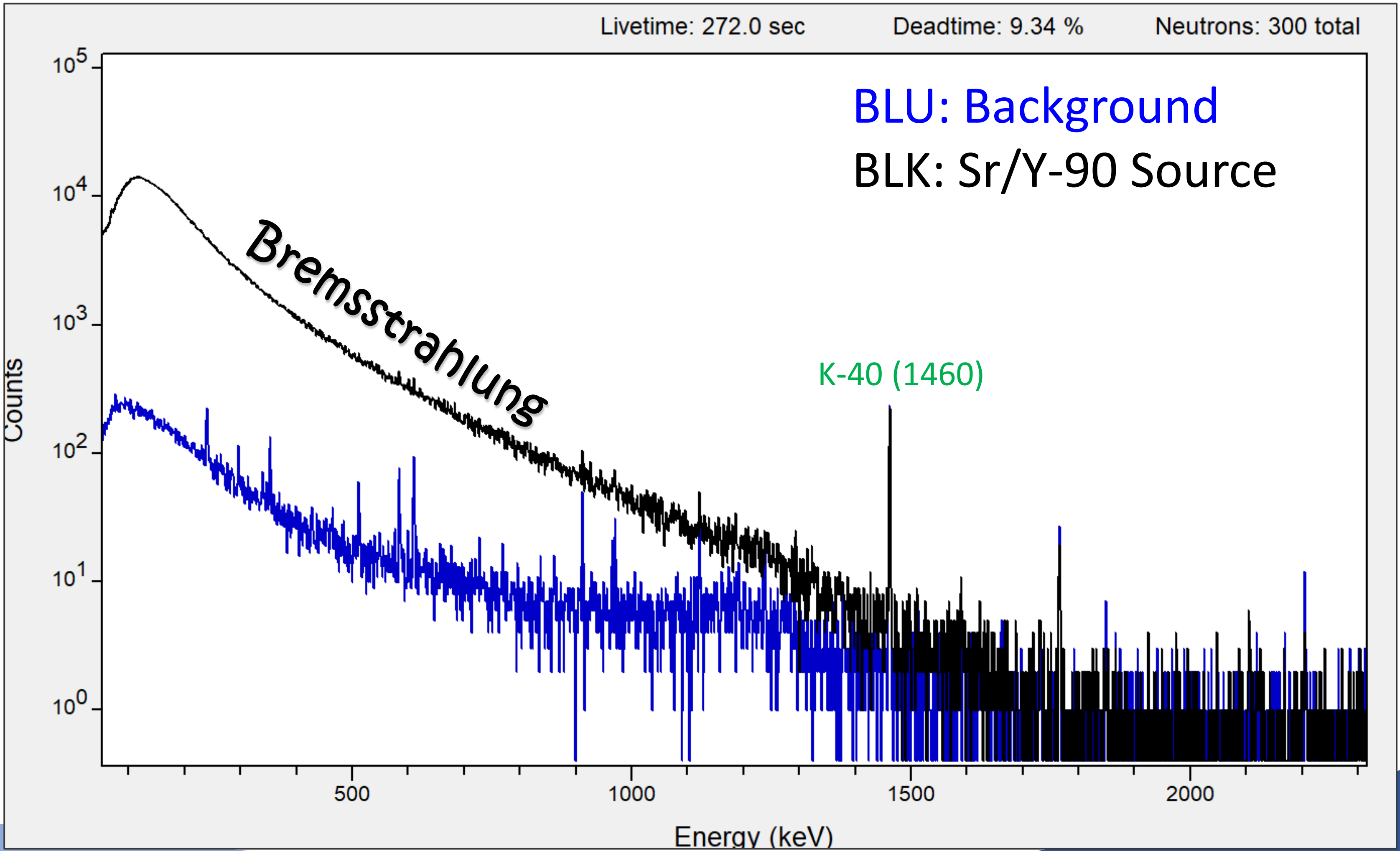
- Neutron-Capture Lines
- Neutron-Inelastic Scattering Lines
- Counts above 2614.5 keV
- Neutron “ski slopes” in germanium





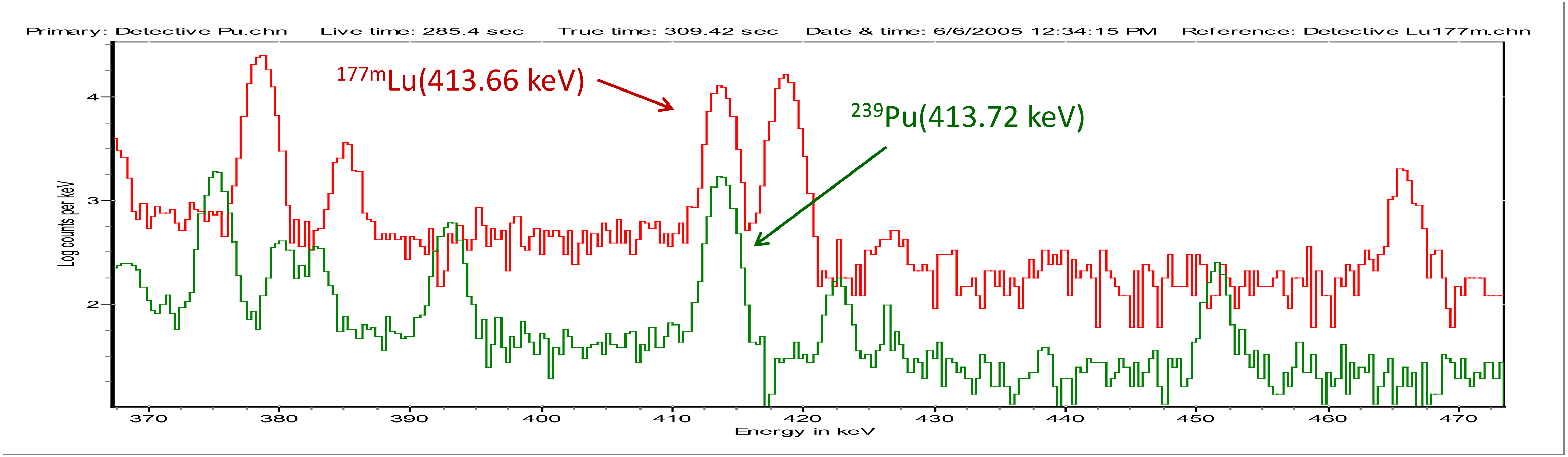
Bremsstrahlung





Degenerate Solutions

- Sometimes (not often) the search result will clearly indicate a single isotope
- Usually, there will be several possible sources



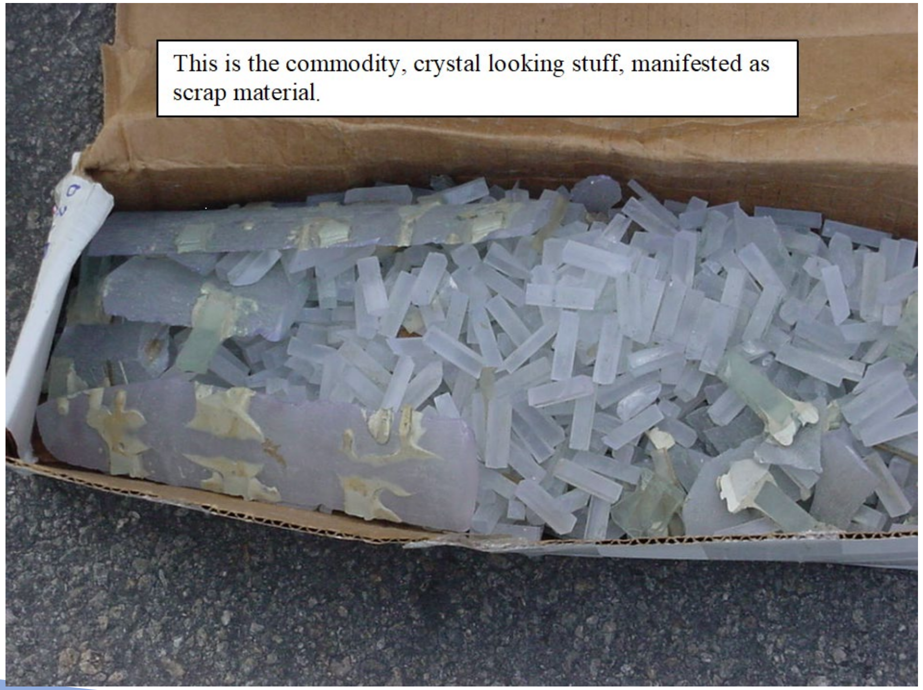
Real-World Example

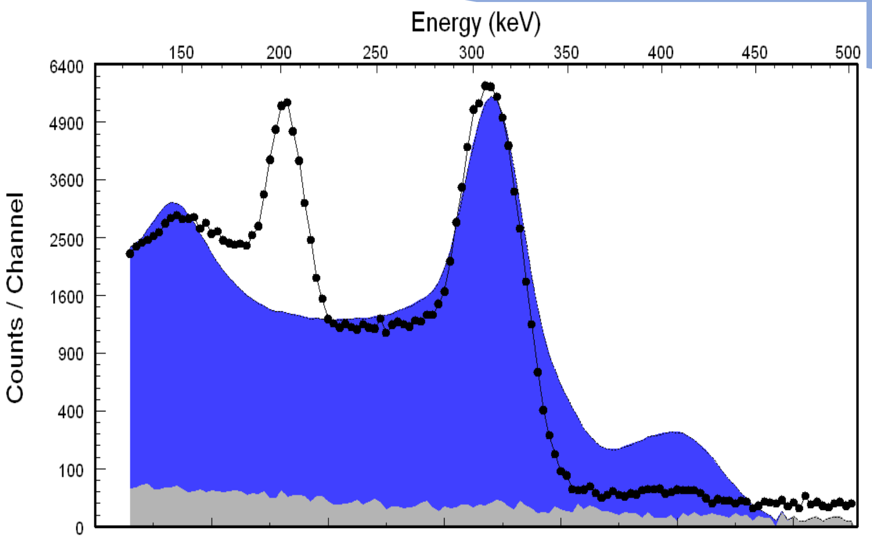
- Shipment seized by U.S. Customs on suspicion of smuggled special nuclear material.

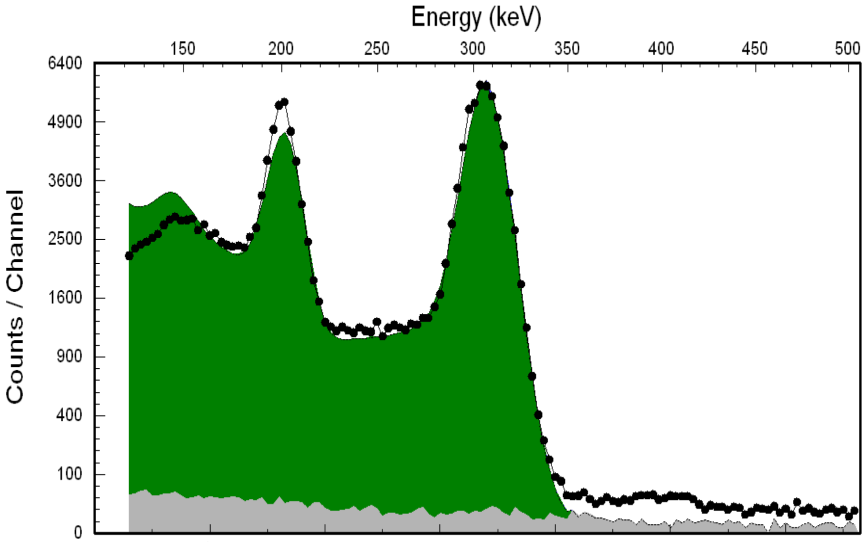
Submission email:
"Per our conversation, see GR-135 file for analysis. In addition to the neptunium-237 we are getting a plutonium-239.(See attached file) sixmin~1.dat"

Real-World Example

- Radioactive boxes contain glass.
- GR-135 reports Np-237 & Pu-239.
- Analysts identified Lu-176, no Np, no Pu







Part 2: Nuclide Quantification

The second step in a gamma-ray analysis of an item is often to determine how much of each gamma-emitting nuclide is present in the item.

3.7E4 Bq

3.7E10 Bq

Nuclear forensics can support the investigative authorities identify the truth about a specific incident by answering the questions what, where, how, when and why an illicit activity occurred and possibly who was involved

Mathematics for Activity Quantification

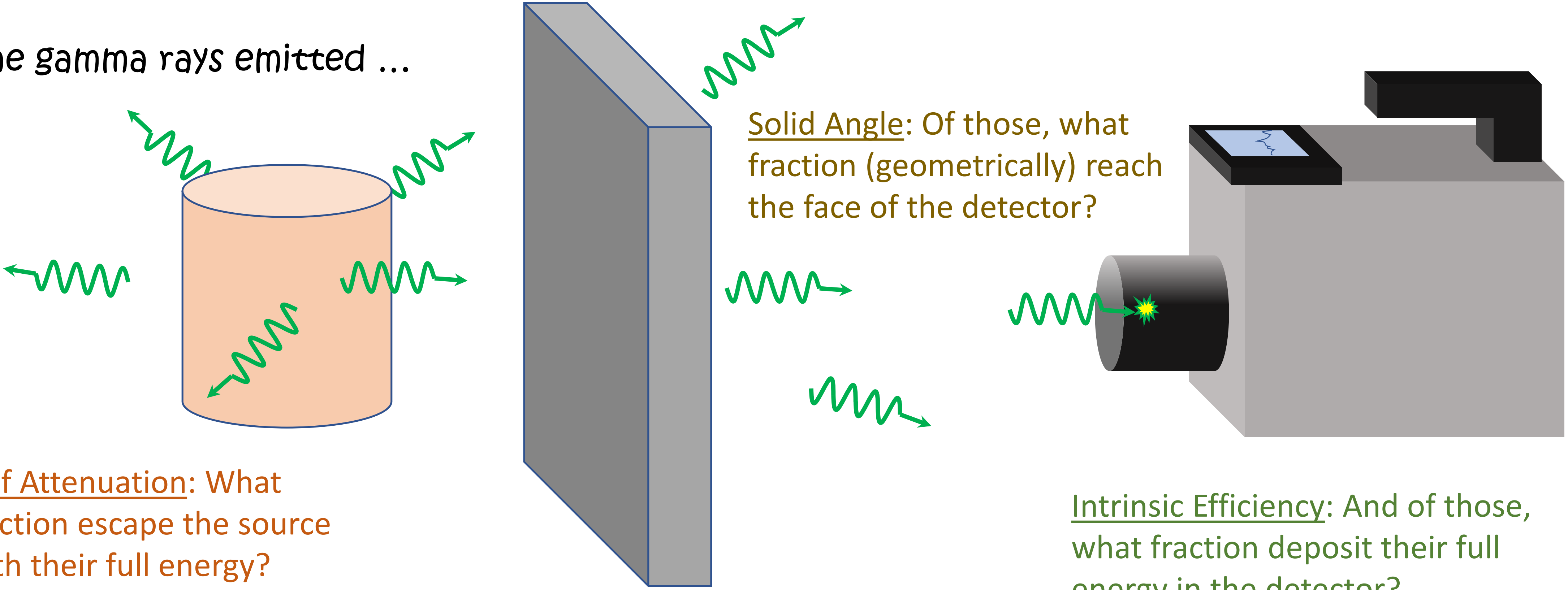
$$Activity = \frac{C(E)}{Y(E)} \cdot \frac{1}{\varepsilon_{Abs}(E)}$$

$$Mass = Activity \cdot \frac{T_{1/2}}{ln2} \cdot \frac{A}{6.022E + 23}$$

C(E): count rate for a specific gamma-ray peak
Y(E): yield (branching ratio) for that gamma ray
ε_{Abs} (E): absolute detection efficiency at that gamma ray energy
$T_{1/2}$: half life of the nuclide emitting that gamma ray
A: atomic mass of this nuclide

Absolute Detection Efficiency

- Of the gamma rays emitted …



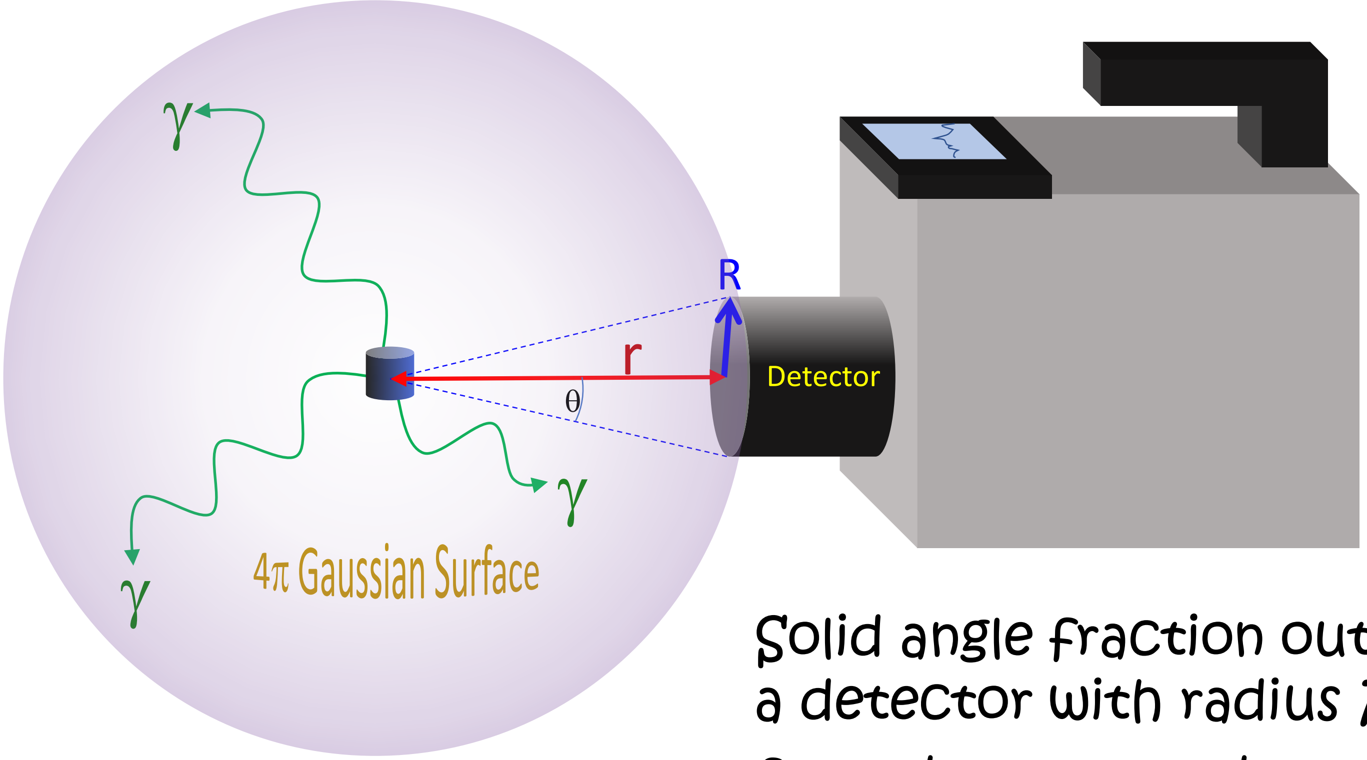
<u>Solid Angle</u>: Of those, what fraction (geometrically) reach the face of the detector?

<u>Self Attenuation</u>: What fraction escape the source with their full energy?

<u>Intrinsic Efficiency</u>: And of those, what fraction deposit their full energy in the detector?

<u>External Attenuation</u>: Of those, what fraction transmit through any intervening material with their full energy?

Solid Angle

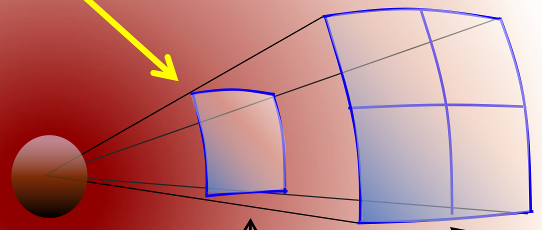


Solid angle fraction out of 4p steradians for a detector with radius *R* at a distance *r* from the source where $\theta = \tan^{-1}(R/r)$:

$$\frac{\Omega}{4\pi} = \frac{1}{2}\left(1 - \cos\theta\right)$$

Inverse-Square Law

Let's say 1 square = the area covered by your detector.



Count rate

$$C \propto \frac{1}{r^2}$$

γ-ray field

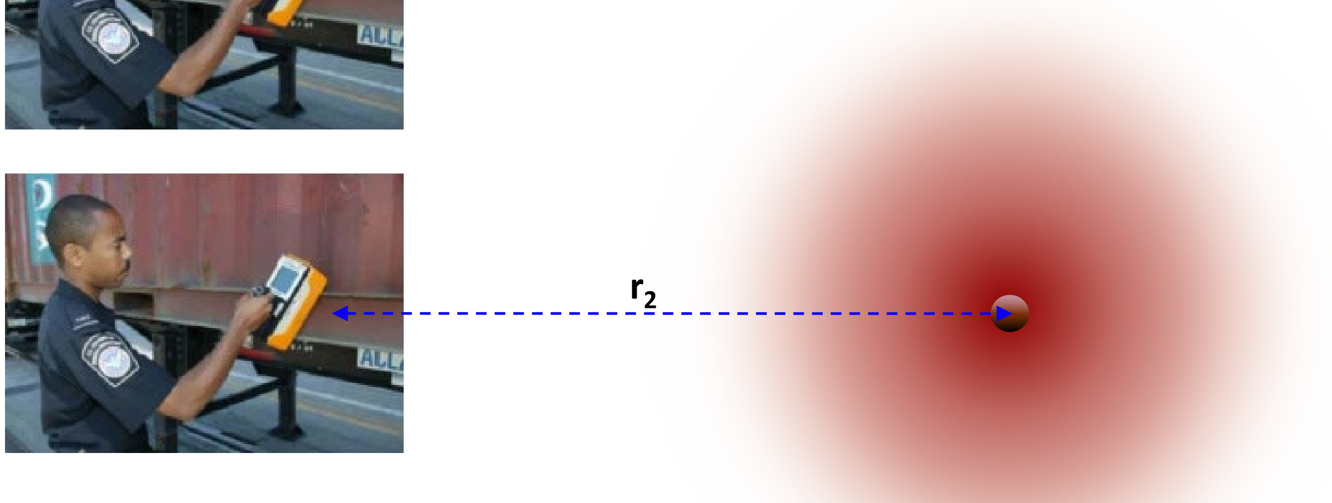
distance = r
area = A
e.g. 400 cps

distance = 2r
area needed for same rate = 4A
Rate for 1 square = 100 cps

If you *double* the distance, the count rate drops by a factor of 4

The Importance of Source-to-Detector Distance

The observed dose rate in these two cases *could* be the same.

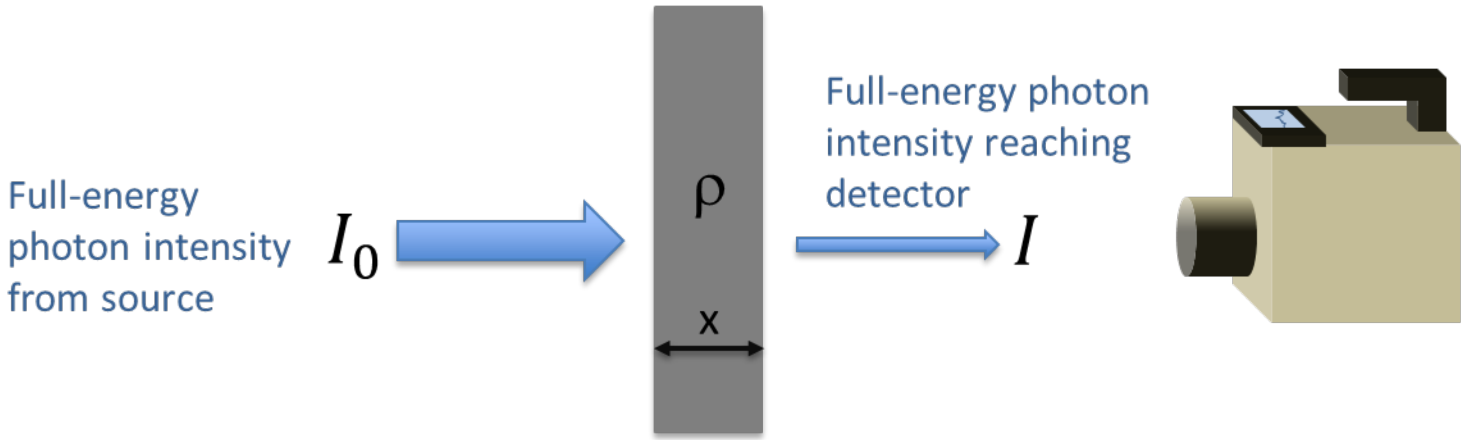


We need to know the source-to-detector distance to calculate the activity or mass of the source.

But the farther source is much more intense!

External Attenuation

- If we know the type and thickness of intervening (external) attenuating materials we can correct for their effect.



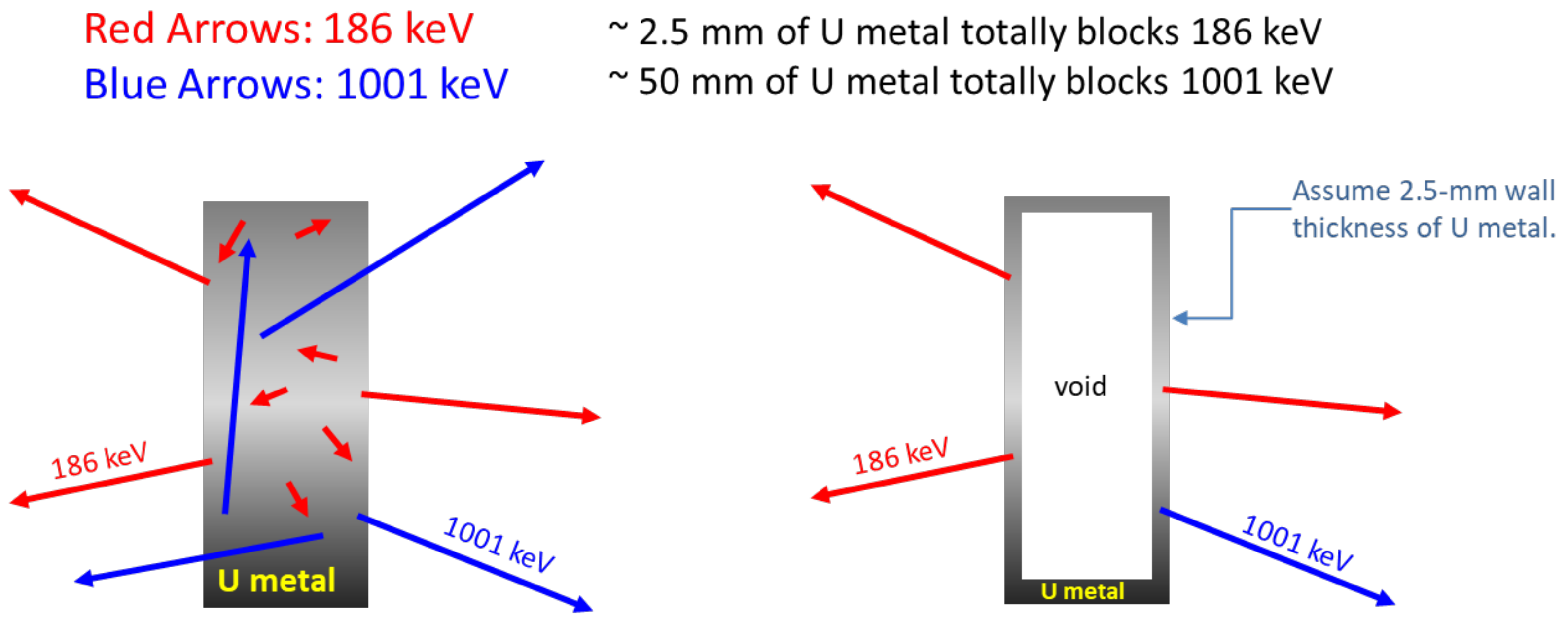
$$I = I_0 e^{-\mu\rho x}$$

We measure an intensity ***I***, but to determine activity of the source we need to know ***I₀***

$$I_0 = I e^{\overbrace{+\mu\rho x}^{\text{correction factor}}}$$

Self Attenuation

- E.g. SNM metal is high-Z, high-ρ material. It is very good at attenuating gamma rays, even those from itself.

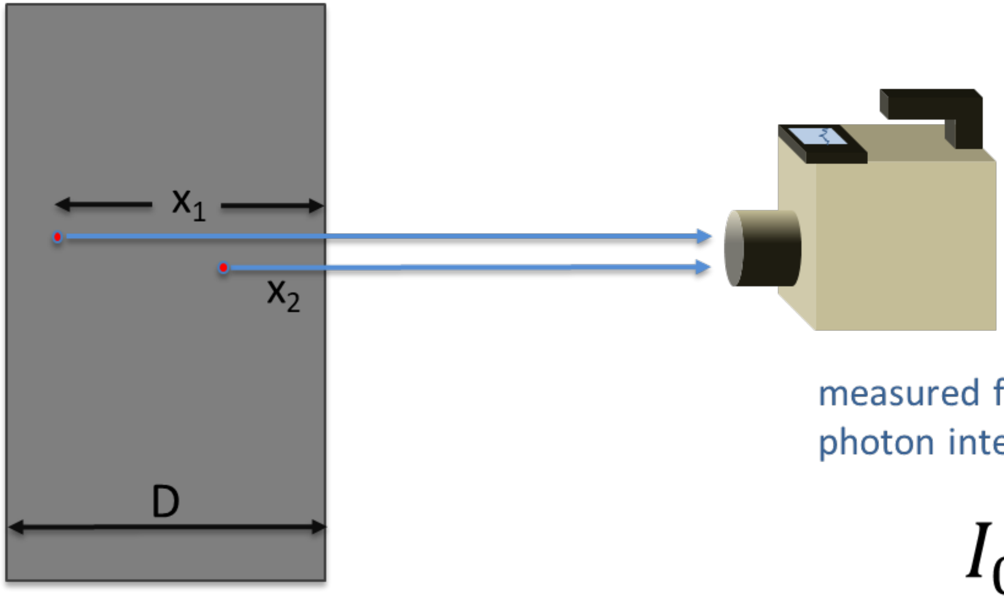


At our detector we see the *same amount of 186-keV* gammas for both cases above. But we see less of the 1001-keV gammas <u>in a relative sense</u> from the thin-walled box than the solid block.

Self Attenuation

- We must integrate over the attenuation experienced at all points in the item at all energies of interest.

In the 1-dimensional case below, photon 1 must transit much more material to escape the item than photon 2.



$$T = e^{-\mu\rho D}$$

correction factor

measured full-energy photon intensity

$$I_0 = I \cdot \underbrace{\left(\frac{-\ln(T)}{1-T}\right)}_{\text{correction factor}}$$

full-energy photon intensity from all atoms in field of view

Relative Efficiency

- General Expression for Peak Area Count Rate: $$\dot{C}(E) = \lambda N \cdot Y(E)\varepsilon_A(E)$$

$\dot{C}(E)$ = count rate at energy E
λ = decay constant
N = number of nuclei
Y(E) = branching ratio (yield) at energy E
$\varepsilon_A(E)$=absolute efficiency at energy E

- Rearrange

$$\varepsilon_A(E) = \left[\frac{1}{\lambda N}\right]\frac{\dot{C}(E)}{Y(E)}$$

- Relative efficiency is proportional to counts and branching ratio

$$\varepsilon_R \propto \frac{C}{Y}$$

Relative Efficiency depends on:
Intrinsic Detector Efficiency

Attenuation

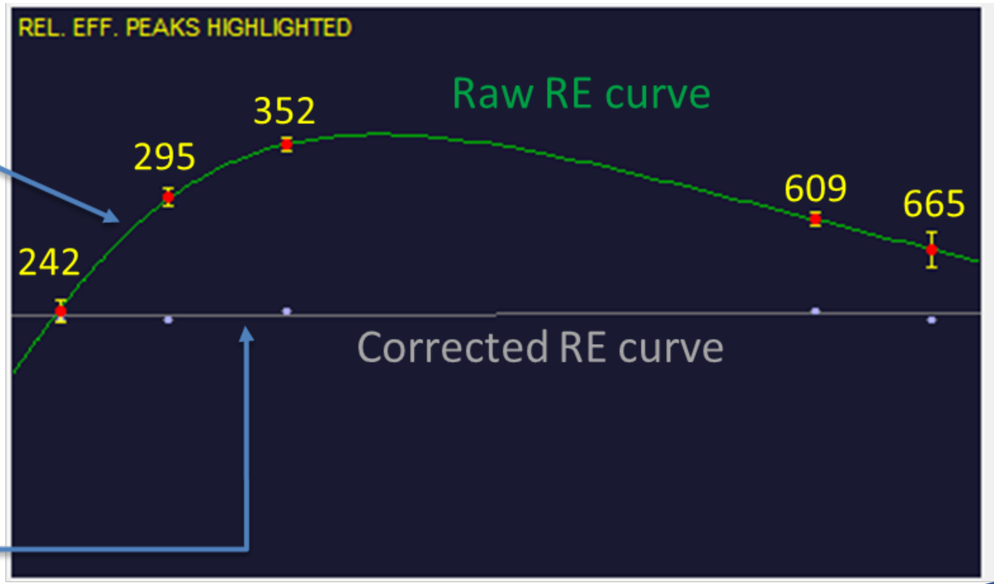
'Flattening' the Relative Efficiency Curve

- A shielded ^{226}Ra source was measured
- Our goal is to vary the amount of Pb to "flatten" the RE curve
  - The corrected data should be linear with energy
  - The slope of the corrected data should be ~zero
- Once we have corrected for RE effects we can calculate the activity

The green curve is a fit to the raw RE data.

At each energy we apply the intrinsic detector efficiency and vary the shielding to obtain the corrected curve

$$RE_{corr} = RE \cdot \frac{\overbrace{e^{+\mu\rho x}}}{\varepsilon_{int}}$$



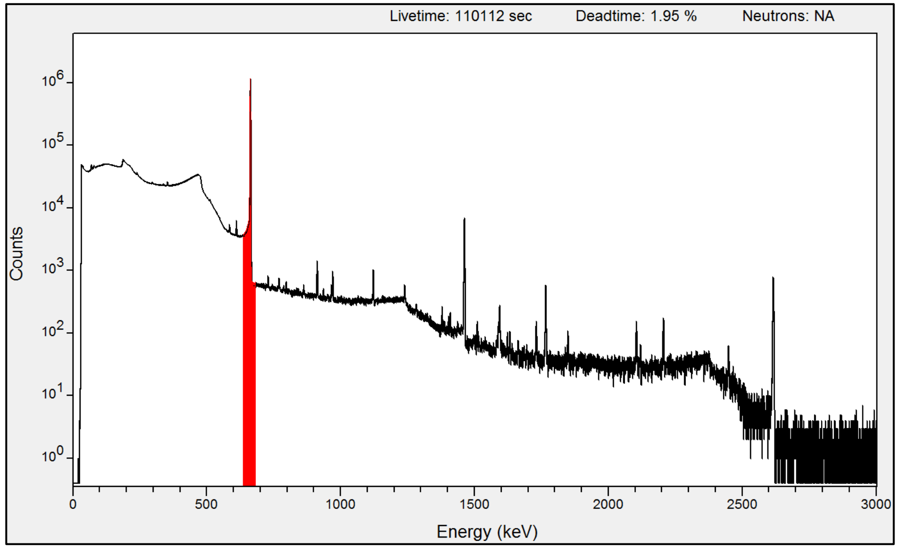
Case 1: Bare Point Source

- Here we measure the activity A of an unshielded ^{137}Cs source
  - Distance: 25 cm
  - Detector: ORTEC Detective (ε_Abs (662 keV)@ 25 cm = 2.7E-04)
  - 662-keV photon yield: 0.851
  - Live Time: 110112 seconds
  - 662-keV Peak Area: 7823347 +- 2851

Count Rate ≅ 71 cps

$$A = \frac{\dot{C}(E)}{Y(E)} \cdot \frac{1}{\varepsilon_{Abs}(E)}$$

$$A = \frac{71}{0.851} \cdot \frac{1}{2.7\times10^{-4}} = 3.1 \times 10^5 \pm 113\ Bq^*$$

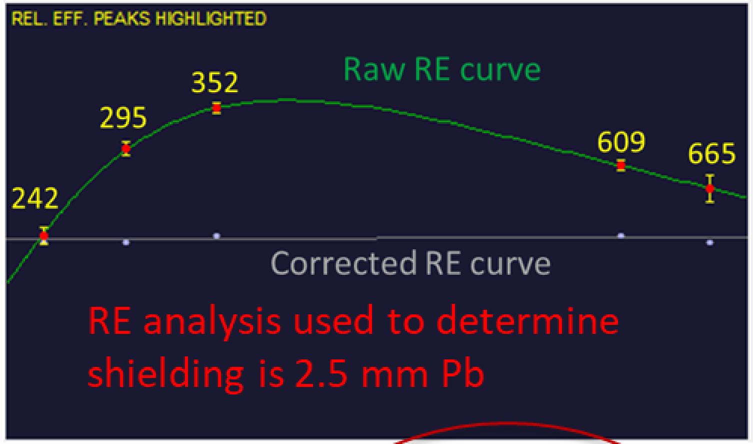


*NOTE: The uncertainty is based on uncertainty in counts. We will address this later.

Case 2: Point Source + External Attenuation

- Here we measure a shielded ^{226}Ra source
  - Distance: 25 cm
  - Detector: ORTEC Detective
  - Live Time: 301 seconds

$$A = \frac{\dot{C}(E)}{Y(E)} \cdot \frac{1}{\varepsilon_{Abs}(E)} \cdot e^{\overbrace{+\mu\rho x}^{\text{Attenuation correction at each energy}}}$$



Error-Weighted Activity = 4.21E6 $\pm$ 2.47E4 Bq

E [keV]	Yield	Counts	Err	RE	Err	DetEff	Mu	e^+upx	RE Corr	Err	Activity [Bq]	Err
242	7.43E-02	11208	171	1.51E+05	2.30E+03	7.30E-04	7.28E+00	6.40E+00	1.32E+09	2.02E+07	4.40E+06	6.71E+04
295	1.93E-01	42051	238	2.18E+05	1.23E+03	5.65E-04	4.72E+00	3.33E+00	1.28E+09	7.25E+06	4.27E+06	2.41E+04
352	3.76E-01	91693	323	2.44E+05	8.60E+02	4.47E-04	3.33E+00	2.33E+00	1.28E+09	4.49E+06	4.24E+06	1.49E+04
609	4.61E-01	98212	322	2.13E+05	6.99E+02	2.28E-04	1.39E+00	1.43E+00	1.33E+09	4.37E+06	4.42E+06	1.45E+04
665	1.46E-02	2862	84	1.96E+05	5.73E+03	2.08E-04	1.24E+00	1.37E+00	1.29E+09	3.77E+07	4.29E+06	1.25E+05

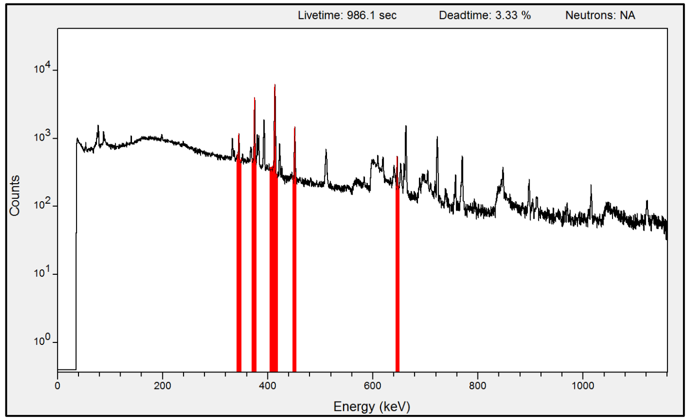
Case 3: Extended Source + External Attenuation

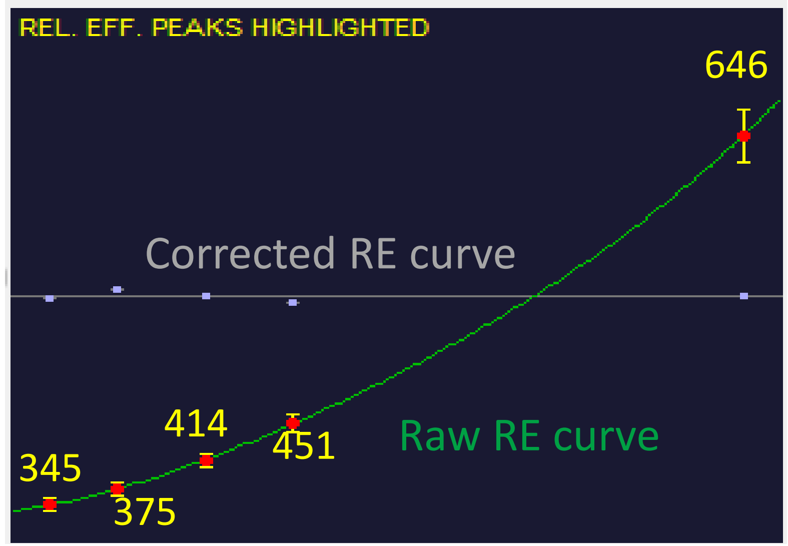
- Here we measure a shielded Pu source
  - Distance: 91 cm
  - Detector: LN2-cooled HPGe
  - Live Time: 986 seconds

$$A = \frac{\dot{C}(E)}{Y(E)} \cdot \frac{1}{\varepsilon_{Abs}(E)} \cdot \underbrace{e^{+\mu\rho x}}_{\text{External Attenuation}} \cdot \underbrace{\left[\frac{-\ln(e^{-\mu\rho x})}{1 - e^{-\mu\rho x}}\right]_{Pu}}_{\text{Self Attenuation}}$$

At each energy we correct for detector efficiency, external attenuation, and now **self attenuation**.

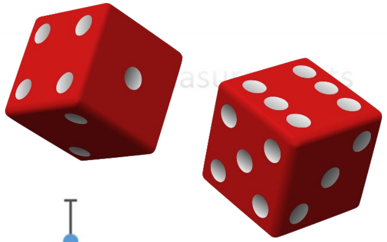
In this example, self attenuation limits our calculated mass to < 20% of the true Pu mass.

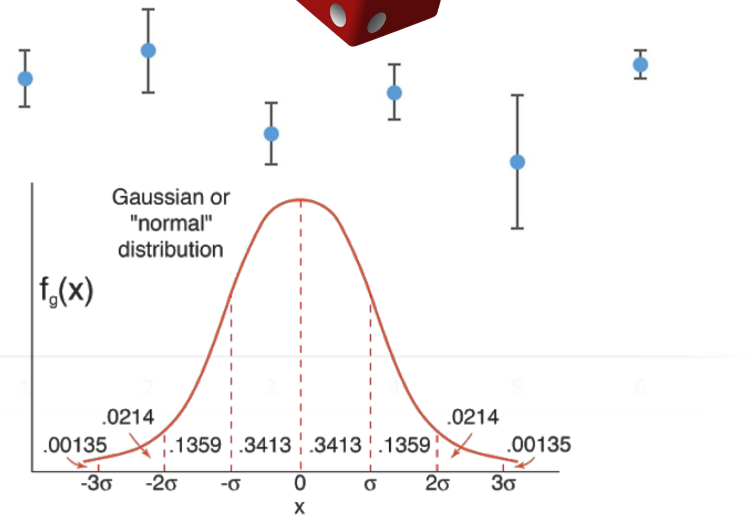




Part 3: Uncertainty Quantification

We must consider the uncertainty involved at each step and how it propagates to the final result.



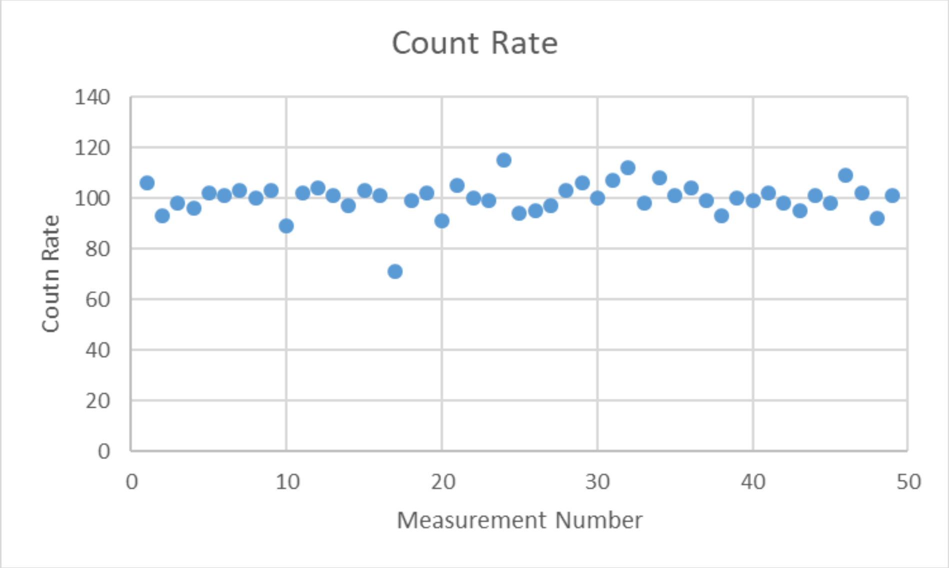


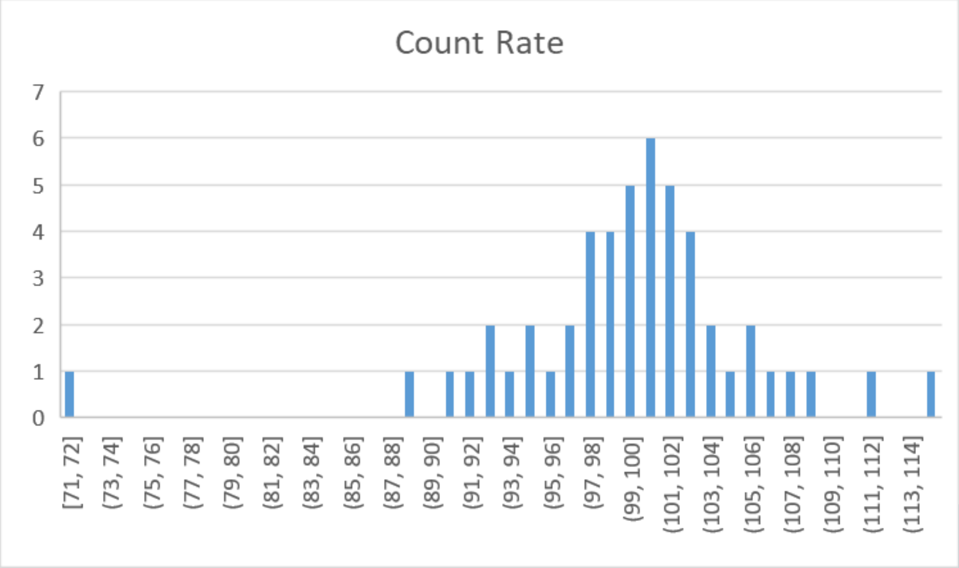
Nuclear forensics can support the investigative authorities identify the truth about a specific incident by answering the questions what, where, how, when and why an illicit activity occurred and possibly who was involved

What is Uncertainty?

- Every measurement result is subject to random and possibly systematic fluctuations that we call ‘uncertainty’ (and sometimes ‘error’)
- Every measurement result should be stated with its associated uncertainty.
  - E.g. Uranium Enrichment: ^{235}U % = 19.9 ± 1.3 %

- If we repeat a measurement many times we will get a <u>distribution</u> of results. The width of that distribution is related to the uncertainty.

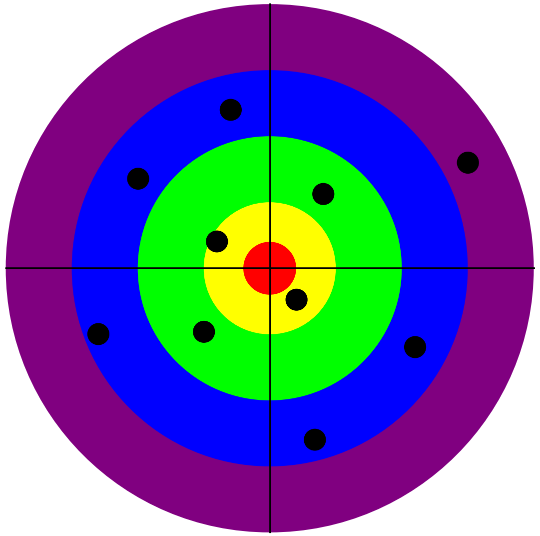


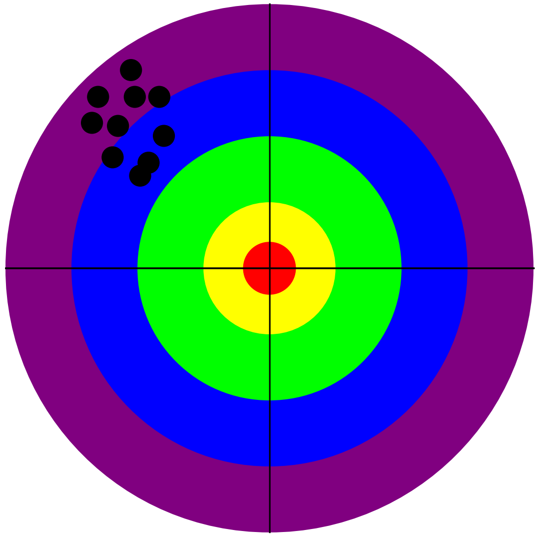


Uncertainty Definitions

- Accuracy: Describes the average value compared to the true value.
- Precision: Describes the variations or dispersion of replicate measurements.
- Bias: Difference between the average value and the true value.

- Random Error: Variable error on replicate measurements.
- Systematic Error: All replicate measurements have the same bias.





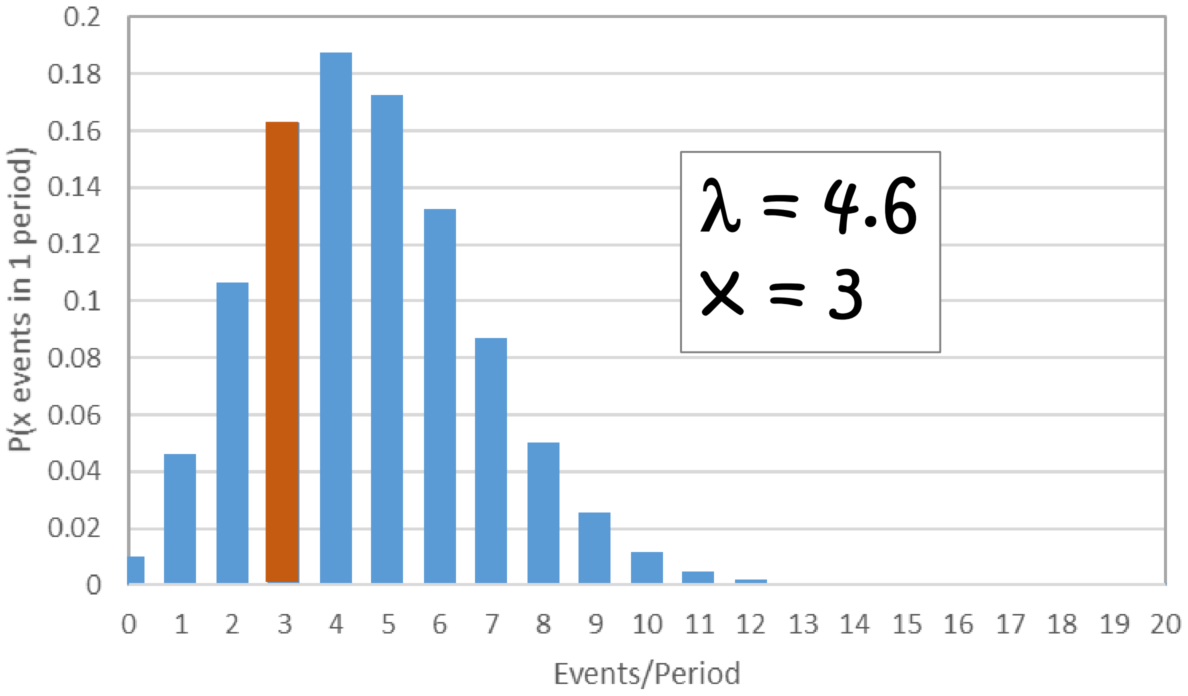
Probability Distributions: Poisson

- Probability of x number of events occurring in a given period of time or space.
- The events occur independently
- The probability that an event occurs does not change with time.

$$P(x) = \frac{\lambda^x e^{-\lambda}}{x!}$$

E.g.: 1 ng of ^{239}Pu → 2.3 decays/sec. What is the probability of 3 decays occurring in a 2-second period?

- 1 period = 2 sec → λ = 4.6 decays/sec.
- x=3 decays
- $P(x=3) = 4.6^3 e^{-\lambda} / 3! = 0.16$

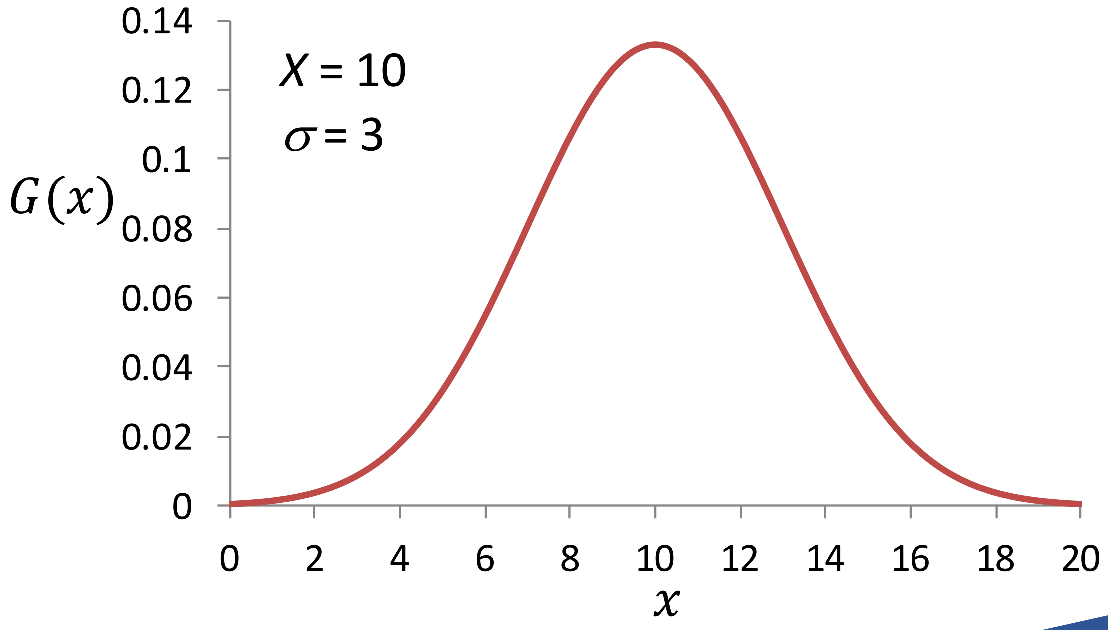


Probability Distributions: Gaussian

X = center of distribution
= average of many measurements

σ = width of distribution
= standard deviation after many measurements
= uncertainty determined from counting statistics from one measurement

$$G(x) = \frac{1}{\sigma\sqrt{2\pi}} e^{-(x-X)^2/2\sigma^2}$$



- For a large number of measurements, X is a good estimate of the true value.

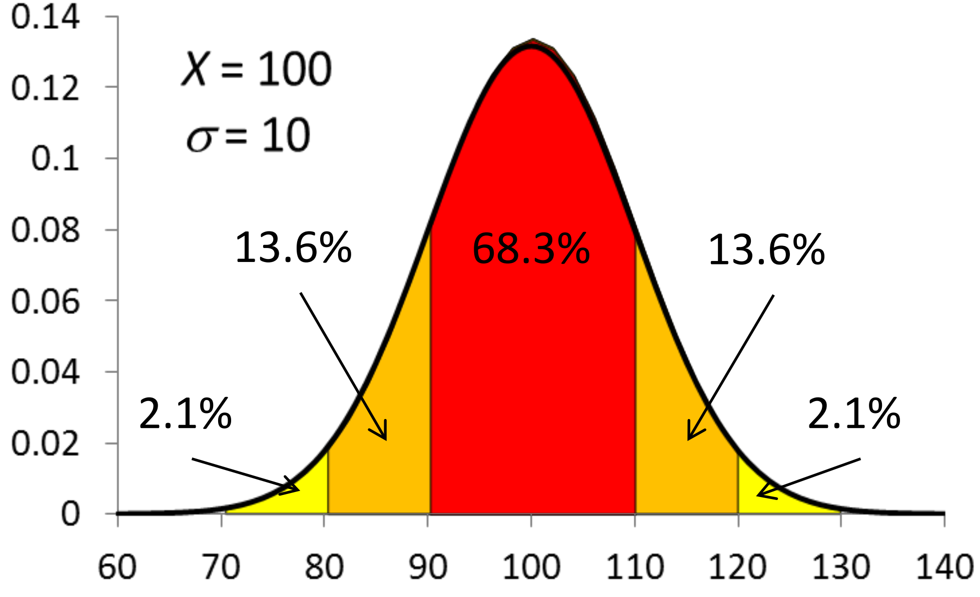
Properties of the Gaussian Distribution

- Probability within:

$$1\,\sigma = \int_{-1\sigma}^{1\sigma} G(x) = 68.3\%$$

$$2\,\sigma = \int_{-2\sigma}^{2\sigma} G(x) = 95.5\%$$

$$3\,\sigma = \int_{-3\sigma}^{3\sigma} G(x) = 99.7\%$$



- How does this relate if only one measurement is made?

- If one measurement is made, there is a 68.3% chance that the true value is within one s and a 95.5% chance that the true value is within two s.

Propagation of Error

The error, σ_f, of a function, $f(x_1, x_2, \cdots, x_i)$, with x_i random variables is propagated using the follow method:

$$\sigma_f{}^2 = \left(\frac{\partial f}{\partial x_1}\sigma_{x_1}\right)^2 + \left(\frac{\partial f}{\partial x_2}\sigma_{x_2}\right)^2 + \cdots + \left(\frac{\partial f}{\partial x_i}\sigma_{x_i}\right)^2$$

For example, if $D = A - B$, then $\sigma_S = \sqrt{(\sigma_A)^2 + (\sigma_B)^2}$

A	12 ± 0.3
B	10 ± 0.4
D	2 ± 0.5
P	120 ± 5.6

if $P = A \times B$, then $\sigma_P = \sqrt{(B\sigma_A)^2 + (A\sigma_B)^2}$

$$\sigma_P = \sqrt{\left(AB\frac{\sigma_A}{A}\right)^2 + \left(AB\frac{\sigma_B}{B}\right)^2}$$

$$\sigma_P = P\sqrt{\left(\frac{\sigma_A}{A}\right)^2 + \left(\frac{\sigma_B}{B}\right)^2}$$

Example: Uncertainty on the Activity of an Unshielded Point Source

$$A = \frac{\dot{C}_N(E)}{Y(E)} \cdot \frac{1}{\varepsilon_{Abs}(E)}$$

Here we stress net count rate with the subscript N.

- Take the simple case where there is only uncertainty on the net count rate $C_N(E)$:
  - $\sigma_Y=0$
  - $\sigma_\varepsilon=0$

$$\sigma_A^2 = \left(\frac{\partial A}{\partial C}\right)^2 \cdot \sigma_{\dot{C}_N}^2 = \left(\frac{1}{Y} \cdot \frac{1}{\varepsilon_{Abs}}\right)^2 \cdot \sigma_{\dot{C}_N}^2$$

$$\sigma_A = \frac{\sigma_{\dot{C}_N}}{Y \cdot \varepsilon_{Abs}}$$

OK so what is $\sigma_{\dot{C}_N}$?

Net Count Rate

$$\dot{C}_N = \dot{C}_T - \dot{C}_B$$

Net Count Rate

Total Count Rate

Background / Continuum Count Rate

$$\dot{C}_N = \frac{C_N}{t_L} = \frac{1}{t_L}\underbrace{(C_T - C_B)}$$

Live Time

Total - Background Counts

Uncertainty on Net Count Rate

Assume there is no uncertainty on the live time

$$\sigma_{\dot{C}_N}^2 = \left(\frac{\partial \dot{C}_N}{\partial t_L}\right)^2 \overset{0}{\cancel{\sigma_{t_L}^2}} + \left(\frac{\partial \dot{C}_N}{\partial C_T}\right)^2 \sigma_{C_T}^2 + \left(\frac{\partial \dot{C}_N}{\partial C_B}\right)^2 \sigma_{C_B}^2$$

$$\sigma_{\dot{C}_N} = \frac{1}{t_L}\sqrt{\sigma_{C_T}^2 + \sigma_{C_B}^2} = \frac{1}{t_L}\sqrt{\underbrace{\sqrt{C_T}^2 + \sqrt{C_B}^2}}$$

$$\sigma_{\dot{C}_N} = \frac{1}{t_L}\sqrt{C_T + C_B}$$

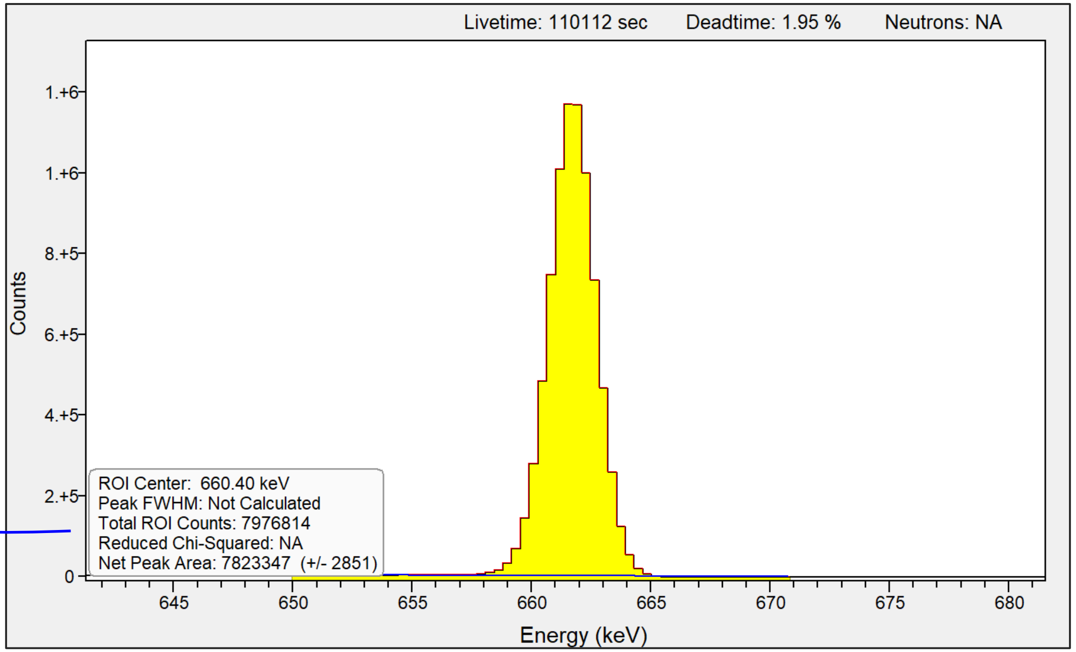
The total and background counts are assumed to follow Poisson statistics

Uncertainty on Activity of a Bare Point Source

$$\sigma_A = \frac{\sigma_{\dot{C}_N}}{Y \cdot \varepsilon_{Abs}} = \frac{\sqrt{C_T + C_B}}{t_L Y \cdot \varepsilon_{Abs}}$$

Using the values from our previous example of an unshielded point source:

$$\sigma_A = \frac{\sqrt{7976814 + 153467}}{110112\ s \cdot 0.851 \cdot 2.7E-4}$$



$$\sigma_A = 113\ Bq$$

Using a 95% confidence interval (2σ):

Our previously-calculated Activity = $3.1 \times 10^5 \pm 226\ Bq$

Questions or Comments?